

RELATIONS BETWEEN STRUCTURE AND MECHANICAL
PROPERTIES OF AUTOCLAVED AERATED CONCRETE

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ABSTRACT

Laboratory specimens of autoclaved aerated concrete were produced under varying conditions, mainly with cement and lime as binders. The type and amount of reaction products, the porosity and the pore size distribution were studied. Shrinkage and compressive strength were measured. The reaction products belonged to the tobermorite group of calcium silicate hydrates and the term crystallinity was defined as the percentage of 11.3 Å tobermorite out of the total amount of calcium silicate hydrates. The shrinkage decreased with increasing crystallinity while the compressive strength increased up to an optimum value. The strength also increased with increasing amounts of hydrates and with decreasing porosity. Other features of the reaction products were indicated by thermal behaviour and micropore size distributions and may have been of importance for the mechanical properties of the material.

On fabrique des échantillons de laboratoire de béton cellulaire en autoclave sous différentes conditions, principalement avec du ciment et de la chaux comme liants. On étudie le genre et la quantité des produits de réaction, la porosité et la répartition du diamètre des pores. On mesure le retrait et la résistance à la compression. Les produits de réaction appartiennent au groupe tobermorite des hydrates silico-calcaires. On définit le terme de 'cristallinité' comme le pourcentage de 11.3 Å tobermorite de la quantité totale des hydrates silico-calcaires. Le retrait diminue avec la cristallinité croissante, tandis que la résistance à la compression augmente jusqu'à une valeur optimale. La résistance augmente aussi avec la quantité croissante des hydrates et avec la porosité décroissante. Le comportement thermique ainsi que la répartition du diamètre des micropores indiquent d'autres caractéristiques des produits de réaction qui peuvent influencer les qualités mécaniques du matériau.

Introduction

Autoclaved aerated concrete is a cellular product in which the binding material consists of calcium silicate hydrates. It can be made from various raw materials such as cement, lime, granulated blast furnace slag, sand and pulverised fuel ash. The cellular structure is normally formed by including fine aluminium powder in the mix. This reacts with lime or other alkaline substances to produce hydrogen, which causes the so called rising of the mix. After the rising, the aerated concrete has to harden for a few hours before it can be cut and autoclaved.

The work reported here has been concerned with the problems of relating the structure of the material to the mechanical properties, viz the shrinkage and the compressive strength. The material structure is characterised by its solid material and its pores. The pores are of two kinds; the macropores, which are the result of the rising of the concrete, and the micropores, which appear in the walls between the macropores. The micropores are the remains of the originally waterfilled space which partly becomes occupied by reaction products during the autoclaving. The material structure can be characterised by the type of reaction products, the amount of reaction products, the porosity and the pore size distribution.

Methods

Laboratory specimens were produced under various conditions including variations in raw materials, additives, mix proportions, densities and autoclaving conditions. In most specimens cement and lime were used as binders and the source of silica was a quartz sand. Alumina containing materials such as granulated blast furnace slag, bauxite and high alumina cement were used as additives. The density of the blocks was made to be about 500 kg/m^3 except for one series where the density was varied between 315 and 265 kg/m^3 . The autoclave pressure was kept constant at 1 MN/m^2 above atmospheric while the autoclaving time was varied. The conditions of production are beyond the scope of this report, since the objective was to find relations between the material structure and the mechanical properties rather than to study the influence of various conditions of production.

The reaction products were studied in various ways, X-ray diffraction being the most important. Films were taken with a Nonius focussing camera using $\text{Cu K}\alpha$ radiation. The films were quantitatively evaluated with a densitometer. By using 10% CaF_2 as internal standard the amounts of unreacted quartz and tobermorite could be determined. Four exposures were taken of each sample and the co-efficient of variation for the relative peak heights was on the average 4% for quartz and 8% for tobermorite. The 4.26 \AA and the 11.3 \AA peaks were used respectively.

The reaction products were also studied by differential thermal analysis (DTA). For a few samples the specific surface was measured by nitrogen adsorption using the BET method and some samples were studied in a transmission electron microscope using the replica technique. The total amount of calcium silicate hydrates was measured by chemical analysis, using Stokes' (1) method.

The porosity was determined with a pycnometer method using water as measuring medium. Very limited studies were made of the macropore size distribution using an image analyser and of the micropore size distribution using a mercury

porosimeter. The shrinkage was measured by the standard method used in Sweden, ie drying of prisms 40 x 40 x 160 mm in 43% relative humidity after three days of water soaking. The compressive strength was also measured by the Swedish standard method, which uses cubes 150 x 150 x 150 mm. The strength results were corrected to be valid at a moisture content of 10% and a density of 500 kg/m³ (except in that series where the density was varied). The corrections were made according to the following empirical formula which is based on experience in the Siporex laboratories:

$$\tau_{\text{corr}} = \tau_{\text{act}} \times \frac{\log_{10} (2466 / (2680 - D_{\text{nom}}))}{(0.87 + (2.02 / (5.7 + f))) \times \log_{10} (2466 / (2680 - D_{\text{act}}))}$$

where τ_{corr} (MPa) is the compressive strength valid at the density D_{nom} (kg/m³), which in this case was chosen as 500 kg/m³, and the moisture content 10%. τ_{act} is the strength actually measured at the density D_{act} (kg/m³) and the moisture content $f\%$.

Type of Reaction Products

X-ray Diffraction

From the XRD results it was found that the main reaction products in all cases belonged to the tobermorite group of calcium silicate hydrates. This group can be subdivided into crystalline, semi-crystalline and near amorphous tobermorites (2). Of the crystalline tobermorites only the 11.3 Å tobermorite was found to a greater or lesser extent in the samples. For simplicity the term tobermorite will be used in the following to denote 11.3 Å tobermorite.

By comparing the relative intensities of the 3.08 Å reflections in the samples with those of the same reflections in a standard preparation of tobermorite, it could be seen that the 3.08 Å peak could not be accounted for by the tobermorite alone. Since the strongest reflection of the semi-crystalline tobermorites is near to 3.08 Å it seems reasonable to assume that the aerated concretes contain a mixture of crystalline and semi-crystalline material. The chemical analysis showed that most samples had a C/S ratio between 0:8 and 1:0, which indicates that the semi-crystalline material can be classified as C-S-H (I).

Thus it can be said that the reaction products consisted of a mixture of tobermorite and C-S-H (I). Normally this would be interpreted as the material being a mechanical mixture of the two components in certain proportions. However, an alternative interpretation is possible. Since the tobermorite and the C-S-H (I) are closely related, differing mainly in crystallinity, one can look upon these materials as end members of a series of materials with varying degrees of crystallinity. A sample which from the XRD-analysis appears to be a mechanical mixture of tobermorite and C-S-H (I) could according to the latter interpretation rather be an intermediate, homogeneous material with a degree of crystallinity somewhere in between those of tobermorite and C-S-H (I). From the XRD results it is not possible to distinguish between the two interpretations. Probably one has to deal with a combination of both cases, ie mechanical mixtures of intermediate materials with varying degrees of crystallinity. Similar reasoning is given in (3).

On the basis of the above reasoning the term crystallinity has been defined as follows:

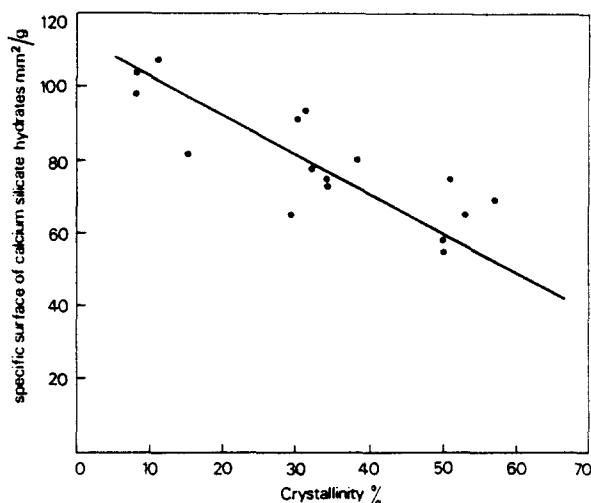


FIG. 1

Relationship between specific surface of the calcium silicate hydrates and the crystallinity.

$\% \text{ Crystallinity} = \% \text{ tobermorite} \times 100 / \% \text{ total calcium silicate hydrates}$
or, if only tobermorite and C-S-H (I) are present:

$$\% \text{ Crystallinity} = \% \text{ tobermorite} \times 100 / (\% \text{ tobermorite} + \% \text{ C-S-H (I)}).$$

The amounts of tobermorite and C-S-H (I) should be considered as equivalent ones, rather than true ones, since according to the above reasoning, one can assume that more than two types of materials may be present

Specific Surface

Since no other hydrate phases than calcium silicate hydrates were detected, the specific surface area of these phases could be calculated from that of the whole material. The relationship between the crystallinity and the specific surface area is shown in Fig 1, which indicates that the less crystalline calcium silicate hydrates have a higher specific surface than well crystallised material.

Differential Thermal Analysis

In almost all the samples, an exotherm somewhat above 800°C appeared in the thermograms. This is most likely due to the formation of β -wollastonite. According to Kalousek (4) pure tobermorite does not give it while Al-substituted tobermorite does. Kalousek also was of the opinion that C-S-H (I) gives an exotherm peak about 800°C (5), but others (6,7) doubted that this is so.

In the present investigation the height of the exotherm peak could be related to the content of C-S-H (I), the latter being calculated as the difference between the total amount of calcium silicate hydrates and the content of tobermorite, since no other calcium silicate hydrate was detected. For cement and lime mixes without additions of alumina containing materials (except of course the cement), the peak height increased linearly with the C-S-H (I) content, thus supporting the view that C-S-H (I) does cause an



FIG. 2

Electron micrograph of aerated concrete.

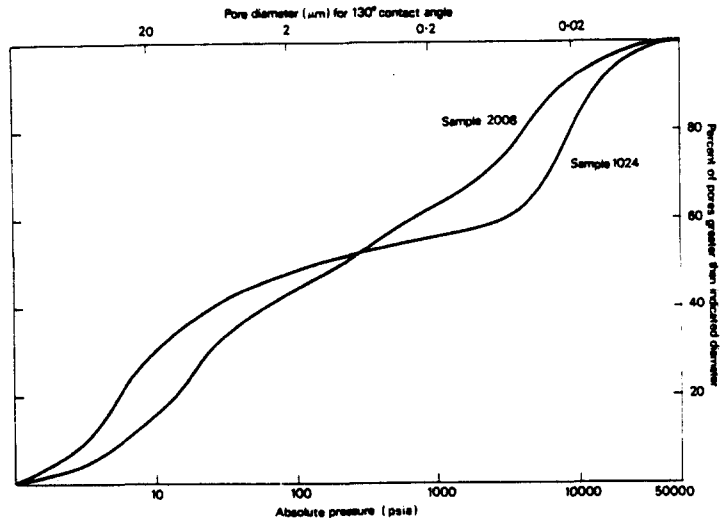


FIG. 3
Micropore size distribution of two aerated concrete samples measured by mercury porosimetry.

exotherm above 800°C. When alumina containing materials were added, however, the peak height increased due to some other substance than C-S-H (I), probably Al-substituted tobermorite as stated in (4). The peak temperature increased with increasing crystallinity but the mixes containing additions of bauxite or high alumina cement were anomalous in this respect, giving higher peak temperatures than the others. In some samples, a small unidentified endothermal effect was also found at about 410°C.

Electron Microscopy and Micropore Size Distribution

Some samples were studied by electron microscopy. Well crystallised plates of tobermorite as well as less crystalline material was seen (Fig 2). The size shape and other features of the plates differed between the different samples. Two samples were studied by mercury porosimetry. The samples showed different size distributions (Fig 3), in spite of the fact that they had very similar XRD patterns, which shows that crystallinity, as defined above, is not sufficient to describe the reaction products.

Relations Between Structure, Shrinkage and Strength

Influence of Crystallinity

The crystallinity was found to have a decisive influence on both shrinkage and strength. In Fig 4 shrinkage has been plotted against crystallinity for mixes based on cement and lime. The figure includes mixes with additions of slag but not those with bauxite or high alumina cement. It also includes mixes with varying porosity although most have the porosity corresponding to a density of about 500 kg/m³.

In the case of the strength, it was found that apart from the crystallinity, the total amount of calcium silicate hydrates and the porosity had an influence. In Fig 5, therefore, this influence has been excluded by only plotting samples with about the same amount of calcium silicate hydrates (50-60%) and the same density (500 kg/m³). There appears to be an optimum crystallinity up to which the strength increases. Some samples deviate considerably from the mean curve, probably because of characteristics of the structure which are not taken into account by the crystallinity.

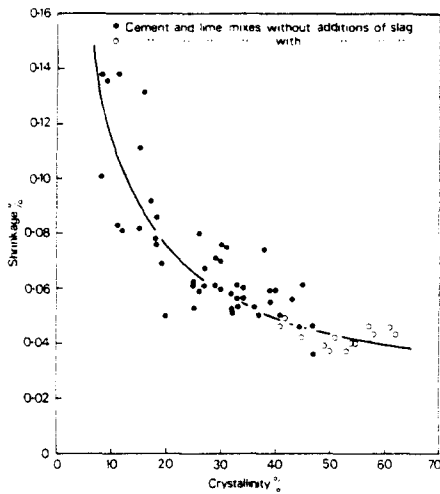


FIG. 4

Relationship between crystallinity and shrinkage for cement and lime mixes without or with additions of slag.

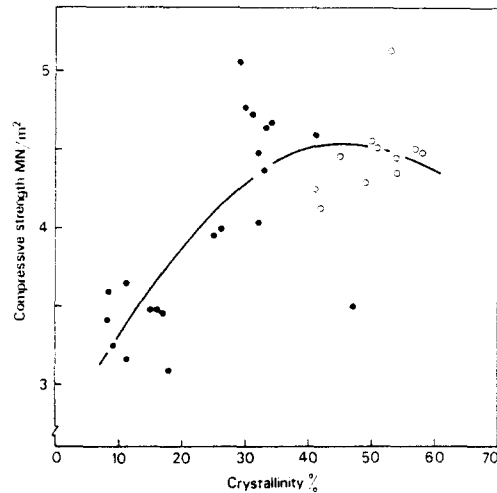


FIG. 5

Relationship between crystallinity and compressive strength for cement and lime mixes without or with additions of slag and a total amount of calcium silicate hydrates of 50-60%. Density 500kg/m³.

Mixes which contained additions of bauxite or high alumina cement showed a systematic deviation from the relations described above. On Fig 6 the relationship between crystallinity and shrinkage is shown for mixes based on cement and lime with additions of bauxite or high alumina cement. For the sake of comparison the relation found in Fig 5 has also been included. It can be seen that the shrinkage is generally lower, sometimes much lower, when these alumina containing substances are added. Also the compressive strength was lower for these mixes. A possible explanation of this behaviour may be found in the DTA results, where considerably higher temperatures of β -wollastonite formation was observed than for other mixes. This must be an indication of changes in the structure of the material.

Influence of the Total Amount of Calcium Silicate Hydrates

Somewhat surprisingly, it was found that the shrinkage was not significantly influenced by the total amount of calcium silicate hydrates when this varied within the limits obtained (38-79%). The compressive strength, however, was influenced by the total amount of hydrates. The combined influence of crystallinity and total amount of calcium silicate hydrates was studied by multiple regression analysis, resulting in the following equation (neglecting some outliers including those with additions of bauxite or high alumina cement):

$$\tau = 0.0386 \cdot T + 0.0831 \cdot C - 0.00087 \cdot C^2 + 0.39$$

where τ = compressive strength (MN/m²), T = total amount of calcium silicate hydrates (%), and C = crystallinity, %.

The coefficient of correlation was 0.90. The equation refers to mixes of density 500 kg/m³.

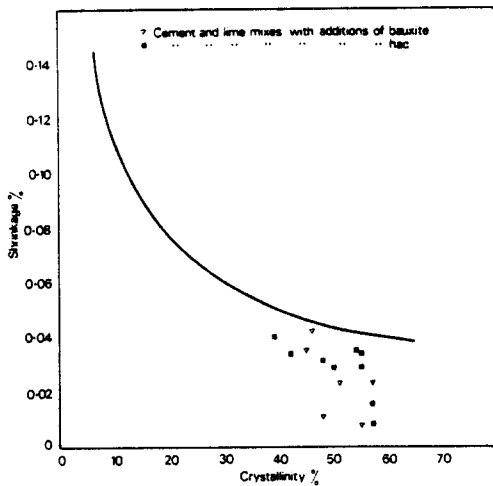


FIG. 6

Relationship between crystallinity and shrinkage for cement and lime mixes with additions of bauxite or high alumina cement.

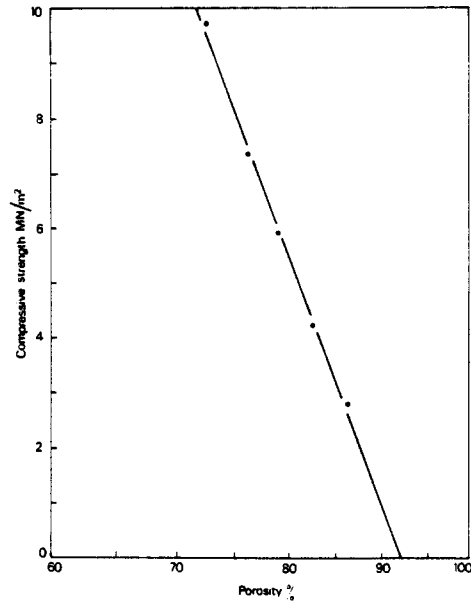


FIG. 7

Relationship between porosity and compressive strength.

Influence of Porosity and Macropore Size Distribution

The most important factor determining the compressive strength was the porosity. In Fig 7 the influence of porosity on the compressive strength is shown for some mixes based on cement and lime without additions, in which only the porosity was varied.

The linear relationship conforms to a Schiller equation indicating that there is a critical porosity of about 92% at which no strength is obtained. The shrinkage increased when the porosity decreased; this can be explained by the fact that the crystallinity decreased at the same time. This is not surprising since a lower porosity means that there is less space in which the crystals can develop. The samples with varying porosity were included in Fig 8.

A very limited study was made of the influence of the macropore size distribution on the compressive strength, but no such influence could be observed.

Conclusions

The reaction products in the autoclaved aerated concretes studied in this investigation belonged to the tobermorite group of calcium silicate hydrates. One important characteristic of these products is what has been called crystallinity, defined as the percentage of 11.3 Å tobermorite out of the total amount of calcium silicate hydrates. Other important factors which influence their behaviour are the micropore size distribution and their chemical importance as indicated by the analysis carried out by DTA.

The shrinkage decreased with increasing crystallinity while the strength increased up to an optimum value after which it decreased. The shrinkage was independent of the total amount of calcium silicate hydrates while the strength increased with increasing amounts.

When bauxite or high alumina cement were added the structure was influenced in such a way as to give lower shrinkage and lower strength.

A linear relationship between the logarithm of the porosity and the compressive strength was found. The shrinkage increased with decreasing porosity due to decreased crystallinity. The macropore size distribution, caused by the aeration, was not found to influence the strength.

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