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EFFECT OF LIGHTWEIGHT FLY ASH AGGREGATE MICROSTRUCTURE ON THE STRENGTH OF CONCRETES

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ABSTRACT

The structure and properties of sintered fly ash lightweight aggregate was modified by heat and polymer treatments to obtain aggregates different in their strength, absorption and pozzolanic activity. These properties of the aggregates were accounted for by changes in their microstructure. The strength of concretes of equal effective water/cement ratio prepared from these aggregates was determined at different ages to resolve the influence of the aggregate properties. It was shown that the strength of the concrete could not be accounted for by the strength of the aggregates only, and it is suggested that the absorption and pozzolanic activity of the aggregates can have an influence on the strength developed. © *1997 Elsevier Science Ltd*

Introduction

In recent years there has been renewed interest in lightweight aggregate concretes, in particular for production of high strength concretes that can be used for bridge construction and other special applications such as off shore structures (1-5). For the production of such high quality concretes there is a need for a better understanding of the mechanisms by which strength is generated in such systems. This is necessary for making the correct choice of aggregates and for developing guidelines for controlled production of lightweight aggregates for this purpose.

In systems of this kind, the aggregate is weaker than the matrix, and is therefore considered to be the weak constituent (6). The mix design concepts are usually based on the production of high strength matrix of low water/binder ratio to compensate for the aggregate weakness. Recent studies of the internal structure of such concretes suggest that the interaction of the paste matrix and the aggregates can be quite complex (7-14) and is different from that of normal aggregate concretes. A variety of processes have been considered to take place between the two phases, associated with special characteristics of such aggregates: rough surface leading to mechanical interlocking, water absorption by the porous aggregates and formation of chemical bond due to the pozzolanic nature of the solid comprising the aggregate. The result of such interactions is an interfacial transition zone which can be quite different from that of normal aggregate concretes; it can be considerably denser depending on the density of the aggregate and its moisture state. The influence of the chemical activity of the aggregate on the structure of this zone is not resolved as in some studies it has been reported to have some influence (10,11), while in others it was reported to have a minor effect if any (9,13).

In view of the significance of the aggregate-matrix interactions in controlling the strength of high strength concretes, it is of importance to resolve such influences in lightweight aggregate concrete systems. The object of the present work was to identify the nature of such interactions and determine their influence on the concrete strength. Thus, the structure of lightweight aggregates was modified to obtain aggregates with different strength, absorption capacity and pozzolanic activity. The strength of concretes produced from such aggregates was evaluated and their microstructure was characterized. On the basis of this systematic data the aggregate-matrix interactions were resolved and their influence on the concrete strength was evaluated.

The first part of this work is presented in this paper. It describes the modification of the aggregates (microstructure and properties) and the resulting influence on the concrete strength. It is shown that the strength cannot be adequately described in terms of the strength of the two phases (aggregate and matrix) and additional factors have to be considered. The second part will deal with the microstructure of these concretes and its correlation with their strength.

Experimental

Aggregate Treatment. Commercial lightweight sintered fly ash aggregate (Lytag, UK) with uniform size grading (maximum size of 6mm and fineness modulus of 4.1) was used as the starting material. The sintering temperature for its production is in the range of 1100-1108C (information provided by the producer).

Commercial aggregate was modified by 4 types of heat treatments and one type of polymer treatment as outlined in Table 1. The table provides a concise description of the treatments and the notations used in this work for each of the aggregates. The heat treatments consisted of heating the aggregates in a laboratory muffle furnace and keeping the desired temperature (1200, 1250, 1300°C) for 30 minutes. The hot aggregate was then removed from the furnace and allowed to cool rapidly in air (cooling rate of about 30°C/min). The aggregate treated at 1300°C was also subjected to a slow cooling rate, by leaving it in the oven that was turned off to cool gradually (cooling rate of about 1°C/min). These treatments exposed the sintered commercial fly ash aggregate to longer retention times than those experienced in the burning process of the coal from which the fly ash was produced, and to

TABLE 1

Treatments and Notation of Aggregates

Lytag	Untreated original aggregate
L1200	Heated to 1200°C, cooled rapidly in air
L1250	Heated to 1250°C, cooled rapidly in air
L1300RC	Heated to 1300°C, cooled rapidly in air
L1300SC	Heated to 1300°C, cooled slowly in oven
polymer	treated with polymer

higher temperatures than those of the sintering process. Thus, mineralogical and microstructural changes were induced.

The polymer treatment consisted of spraying the aggregate with a surface active polymeric material (dimethylhydrosiloxan) in a fluidized bed apparatus in which the aggregate was floated over a spraying stream of the polymer.

Composition and Chemical Activity. The mineralogical composition of the aggregates was estimated by means of X ray diffraction. Petrographic tests of the glassy phase in the aggregates was carried out by means of polarizing microscope equipped with 98 immersion liquids with different coefficients of reflection. The coefficient of the glass reflection can be related to its alkaline or acidic nature (15).

The pozzolanic activity of the external shell of the aggregates was determined by evaluating the calcium hydroxide (CH) content of mixes of ground aggregate shell and cement. The external shell (~0.5mm in thickness) was ground to pass 0.074mm sieve. The powder obtained was mixed with Portland cement (1:9 ratio) and water at a water/solids ratio of 0.35. Cylindrical specimens of 16mm height and diameter were prepared and cured at 20°C water for 28 days, and thereafter in 60°C water up to a total of 90 days. The content of CH was determined at 1,7,28,56 and 90 days by a thermal gravimetric method. The CH content was compared with that of a reference paste of Portland cement only, and from the difference the pozzolanic activity could be estimated. The CH content was calculated as weight per unit weight of the original cement. For that purpose the non evaporable water contents of the cement, the ground aggregate shell and the hydrated samples were determined.

Physical Characteristics. The density and the porosities of the aggregates were determined by means of measuring the weights of oven dry aggregates, vacuum water saturated aggregates and water immersed aggregates. The bulk density of the oven dry aggregate was calculated, as well as the total porosity accessible to water. Pore structure was also determined by mercury porosimetry, with pressure up to 410MPa. The density of the solid phase in the aggregates was calculated from the oven dried bulk densities and the total pore volume. It was found that the mercury porosimetry gave a better estimate of the total porosity, and therefore its pore volume data was used to calculate the density of the solid in the aggregate.

Absorption and rates of water absorption curves were determined by monitoring the change in weight of aggregates immersed in water without vacuuming. This represents the absorption process that takes place in the mixing of lightweight aggregate concrete. Total absorption was determined in vacuum conditions to eliminate entrapped air.

Aggregate Crushing Strength. An estimate for the mechanical quality of the aggregates was obtained by a crushing strength test reported in reference (16). Oven-dried sample of the aggregates was placed in a steel cylinder with internal diameter of 57mm and height of 87mm. The aggregates were filled up to an upper incision mark in the cylinder and afterwards covered with a steel puncheon that was pressed until the upper level of the aggregate was reduced by a prescribed distance. The crushing strength value was calculate in stress units as the ratio between the load and the cross section area of the cylinder.

Lightweight Aggregate Concretes. The lightweight aggregate concretes were all prepared with an effective water/cement ratio of 0.40, after allowing absorption by the aggregates for the first 30 minutes. This was achieved by soaking the aggregates in water for 30 minutes prior to mixing and then rinsing to remove the excess water. The aggregates consisted of a mix of 51% lightweight aggregate and 49% of graded fine aggregate with fineness modulus of 1.95 (0.352 and 0.305 m^3 of lightweight and fine aggregate, respectively per m^3 of concrete). The ratio between the two aggregates was chosen to provide optimal dense grading and workability, based on calculations of Fuller curves followed by trial mixes. The contents of Portland cement and water (in excess of that absorbed at 30 minutes) were 440kg/m^3 and 178kg/m^3, respectively. The Portland cement was a product of Nesher Cement Industries, Israel, equivalent to ASTM Type I. The concretes were cast as 70mm cubes, demolded after one day and continuously cured in lime-water until testing.

Results and Discussion

Composition and Activity of Aggregates. The overall chemical composition of the original aggregate is provided in Table 2.

The X-ray diffraction patterns in Fig. 1 show that the original aggregate contains several crystalline phases: quartz, mullite, anorthite and hematite. The hump in the X-ray diffraction pattern is characteristic of a glassy phase. It can be seen that the increase in the temperature of treatment was associated with a marked reduction in the quartz peak, with only mild increase in the mullite peak. A semi-quantitative estimate of these changes (not shown here) suggest that the quantity of quartz being reduced is greater than that consumed in the formation of other crystalline phases. This implies that the part of the reduction in quartz content is accompanied by formation of additional glassy phase.

Evidence for the increase in glass content can be obtained from the pozzolanic activity test. The CH curves of the mixes with Portland cement and ground shell of all the lightweight aggregates peaked at about 28 days and than declined, in contrast to the reference paste where the CH increased continuously. The decline was greater for the heat treated aggregates, and it increased with the heat treatment temperature. This reduction in CH content with age in the higher temperature treated aggregates is a clear indication of higher pozzolanic activity. To quantify the CH curves (not shown here) in terms of a single parameter, a pozzolanic activity coefficient was defined, which is the difference between the CH content in the reference paste (i.e. the paste without ground aggregate filler) at a particular age and the CH content in the paste with the ground aggregate filler at the same age, relative to the CH content of the reference paste. These coefficients were calculated for pastes at 56 and 90 days, at which time sufficient pozzolanic reaction could have occurred. The values plotted in Fig. 2 clearly indicate increase in the pozzolanic activity with increase in the heat treatment temperature. At 1300°C the pozzolanic activity was highest, as expected. It should be noted that at this temperature even the slowly cooled aggregate showed higher pozzolanic activity relative to all the other aggregates treated at lower temperatures and cooled rapidly.

TABLE 2

Chemical Composition of the Lytag Aggregate

Compound	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O+Na_2O
Content, %wt.	63.1	30.9	4.18	0.72	0.25	0.36	0.40

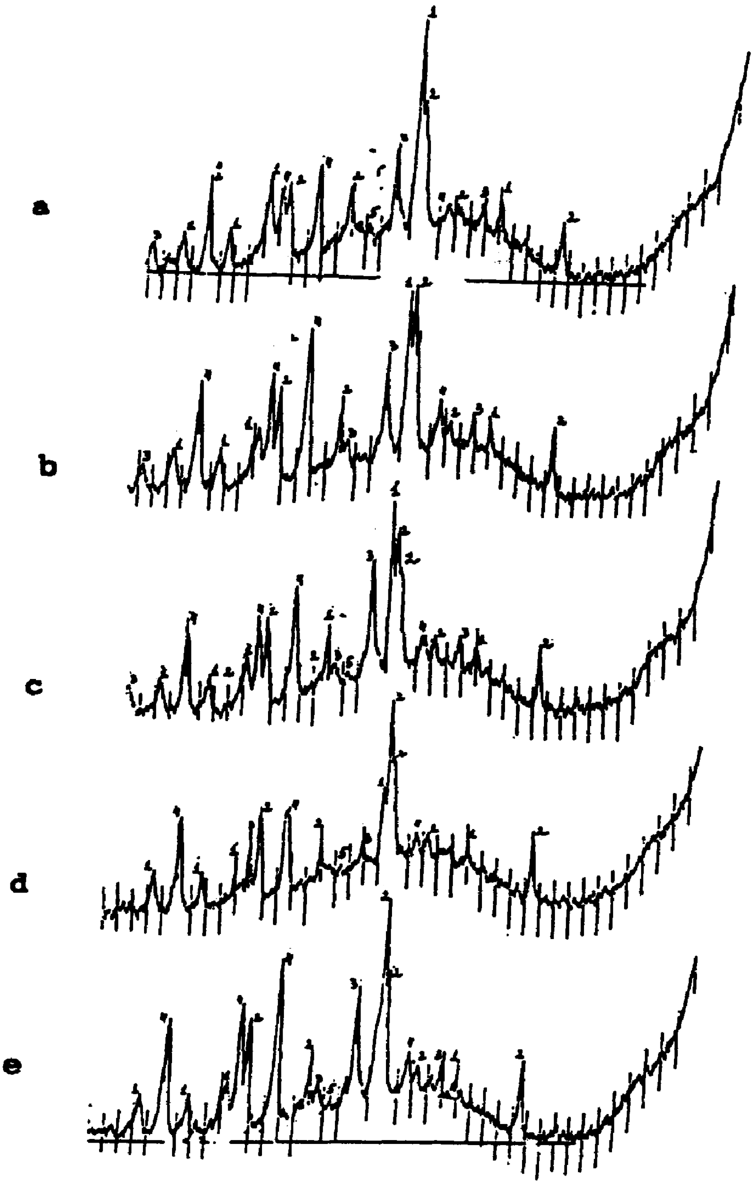

FIG. 1.
X-ray diffraction curves of lightweight aggregates: (a) untreated Lytag, (b) L1200, (c) L1250, (d) L1300RC, (e) L1300SC. Characteristic peaks: 1-quartz, 2-mullite, 3-anorthite, 4-hemathite, 5-diopside.

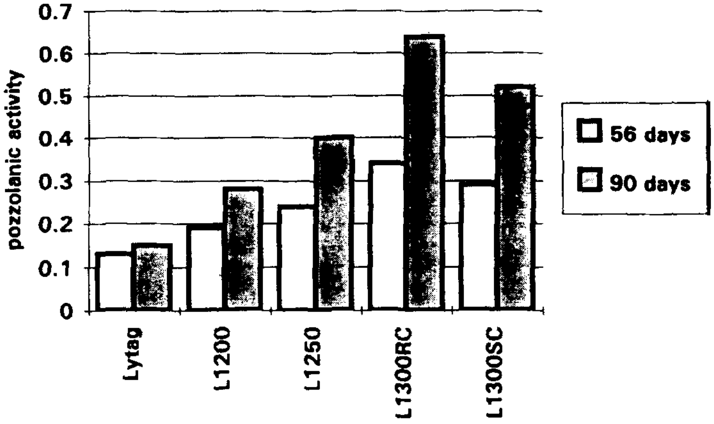

FIG. 2.
Pozzolanic activity coefficients of the external shell portion of the lightweight aggregates.

The enhanced pozzolanic activity at 1300°C treatment may also be associated with the nature of the glassy phase and not necessarily with its quantity only, which was not measured directly in this work. The increase in the temperature of treatment was accompanied by observations of increase in the coefficient of refraction in the petrographic test to values above 1.700. This is a characteristic of a glass with greater tendency for reactivity in basic environment (15).

The mineralogical changes due to the heat treatments are sensitive to the alkali content of the glass. Therefore, different trends may be obtained with fly ash of different compositions.

Microstructure. SEM observations at low magnifications (Fig. 3) shows that in all the aggregates distinction could be made between the core and external shell, ~500-1000μm thick. Large pores bigger than 10μm are usually located in the core, whereas the pores in the shell are smaller. At higher magnifications (Fig. 4) it is possible to see a more uniform distribution

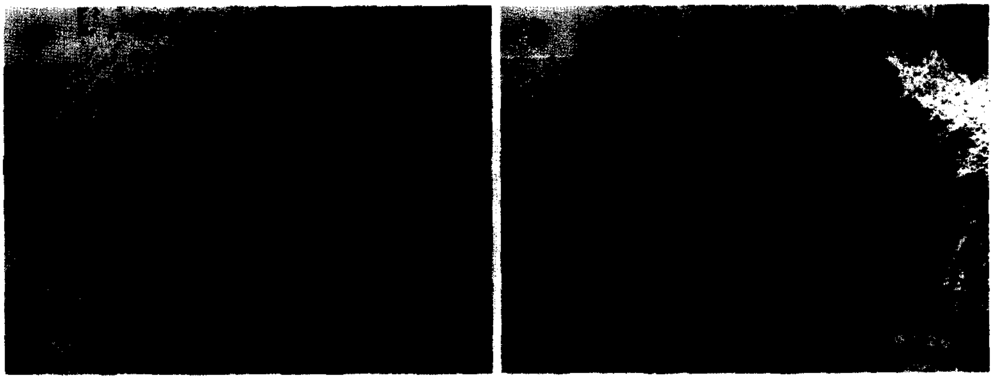

FIG. 3.
SEM observations of fractured lightweight aggregate surfaces: (a) untreated Lytag, (b) L1200.

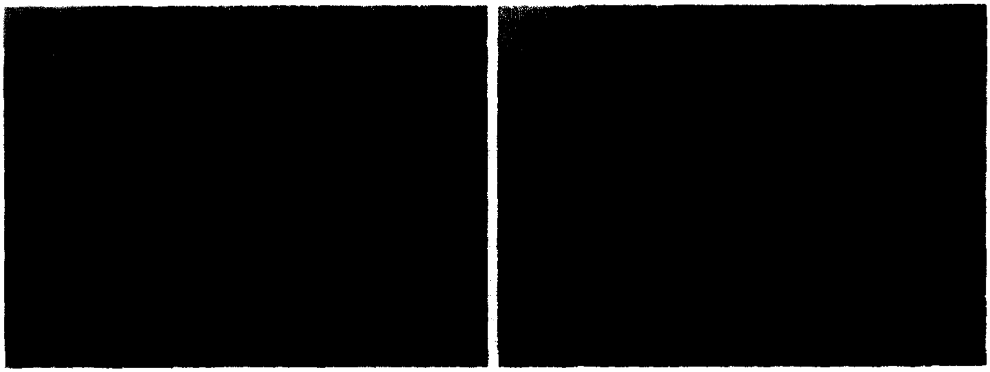

FIG. 4.
SEM observations of fractured lightweight aggregate surfaces: (a) L1200, (b) L1300.

of smaller pores in the higher temperature treated aggregate (1300°C vs. 1200°C). Observations of the external shell of the aggregates clearly show the particulate nature of the sintered fly ash particles in the untreated Lytag aggregate (Fig. 5a) whereas in the heat treated aggregate they become more fused together (similar to the observations in the fractured surface in Fig. 4a). In the aggregate treated with the polymer the spaces between the sintered fly ash particles seem to have become impregnated and their structure can no longer be resolved (Fig. 5b).

Porosity, Density and Absorption. Results of total porosities measured by water saturation and mercury porosimetry are presented in Fig. 6. Generally the heat and polymer treatments reduced porosity in both types of measurements. The relative reduction however is greater in the water saturation values. In all of the aggregates the mercury porosimetry pore volume values were greater than those of the water saturation. This trend is somewhat surprising as usually water porosities are higher due to the more efficient penetration of this liquid into

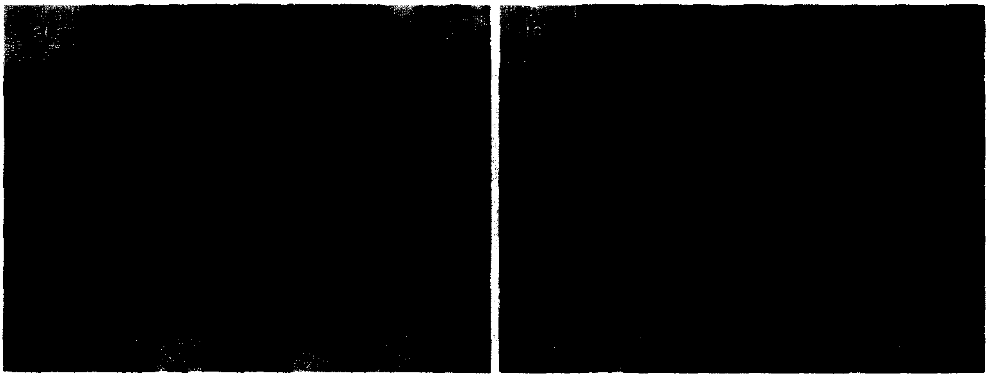

FIG. 5.
SEM observations of the outer surface of aggregates: (a) untreated Lytag, (b) polymer treated.

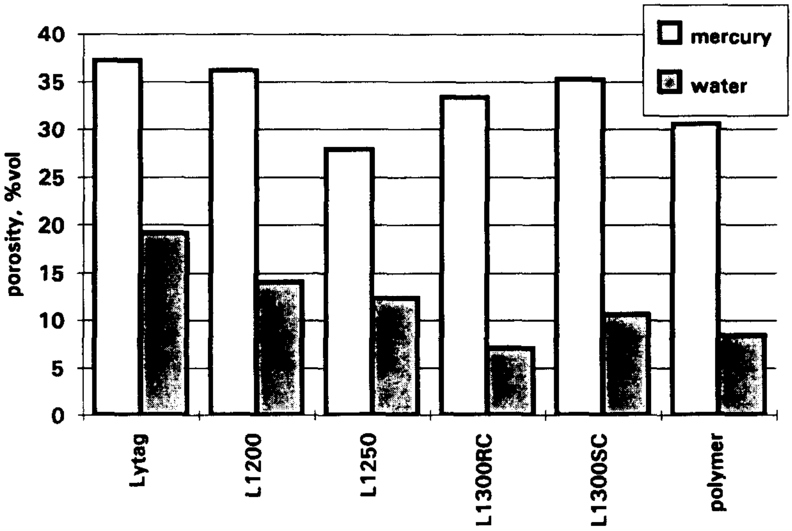

FIG. 6.
Total water and mercury measured porosities of the lightweight aggregates.

smaller pores. The different trend observed here may suggest the presence of a relatively large volume of closed pores which can not be penetrated by water but could be penetrated by mercury, due to rupture of pore walls under the high mercury pressure. Thus, it can be assumed that the mercury porosimetry reflects the total porosity in these systems, whereas the water saturation porosity is a better measure of the open porosity. In terms of the physical and mechanical properties of the aggregates, one would expect that the total porosity (mercury porosimetry test data in this case) would correlate better with the strength of the aggregates while the open porosity (water saturation data in this case) would correlate better with the absorption characteristics of the aggregates. Both properties, strength and absorption, are of prime importance in controlling the behavior of the lightweight aggregate concrete.

The reduction in the porosity of the aggregate might be expected to be accompanied by an increase in the bulk density. However this is not always the case, as seen in Fig. 7a, which shows somewhat erratic trends. However, when considering the changes in bulk density, two factors should be taken into account: the changes in porosity and the changes in the density of the solid in the porous aggregate. The latter values can be calculated based on the values of total porosity (by the mercury porosimetry test) and the bulk density (Fig. 7b). The change in the density of the solids in Fig. 7b shows consistent trends: (i) It reduces with increase in the temperature of heat treatment, which can be accounted for by transformation of the crystalline products to more glassy ones, consistent with the data of X-ray diffraction and pozzolanic activity values presented previously; (ii) For the same temperature of heat treatment, 1300°C, the rapid cooling resulted in lower solid density compared to the slow cooling (1300RC vs. 1300SC in Fig. 7b); (iii) The lower solid density of the polymer treated aggregate relative to the original one (polymer vs. Lytag in Fig. 7b) can be attributed to the presence of a solid which is essentially a mixture of higher density inorganic material and lower density polymeric material, consistent with the SEM micrographs presented previously.

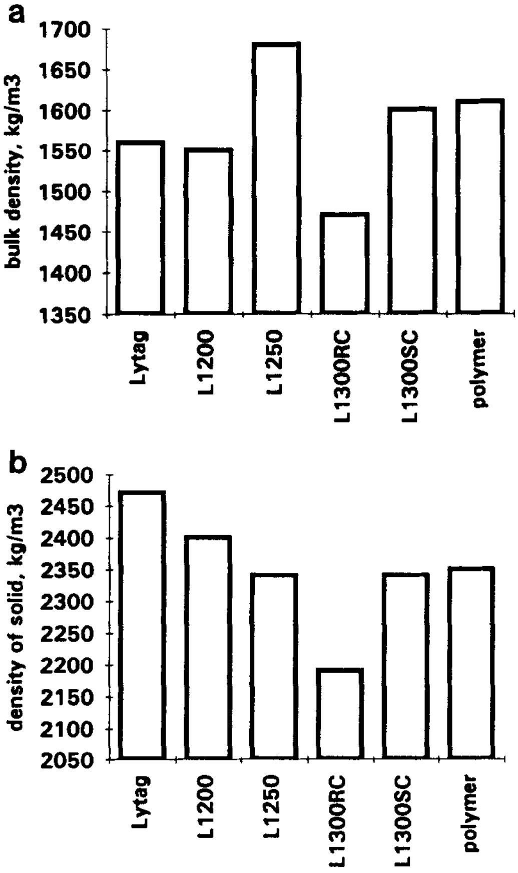

FIG. 7.

Densities of the lightweight aggregates: (a) bulk densities, (b) densities of the solid material comprising the aggregate.

Thus the overall reduction in porosity which should lead to larger bulk density of the porous aggregate is offset by the reduction in the solid density induced by changes in the composition of the solid in the various treatments. Similar arguments may be applied to account for the changes in porosity observed in Fig. 6: generally, increase in the temperature of heat treatment should result in greater fusion and sintering leading to reduction in open porosity; this is indeed the overall trend in Fig. 6. However the reduction in porosity may not be as high as expected by fusion and sintering considerations, due to increase in the volume of the solid which is a consequence of the reduction in its density. This may account for the higher total porosity values of the 1300°C treated aggregates relative to the 1250°C aggregate in Fig. 6.

The consistent trends in Fig. 6 with respect to the change in the open porosity (water saturation values) and the much greater relative decrease in these values, suggest that this porosity, which reflects the structure of the aggregate shell, is influenced to a greater degree by the various treatments, relative to the core of the aggregate. This is in agreement with the absorption data presented in Table 3. The trends of reduced absorption and absorption rate

TABLE 3

Water Absorption at 14 Days

Aggregate type	Lytag	L1200	L1250	L1300RC	L1300SC	polymer
Absorption, %vol.	14.5	10.6	9.7	5.0	9.2	3.8

Remark: About 70% of the 14 days absorption was obtained in 1 day, with the rates of absorption roughly proportional to the total absorption.

with increase in temperature of heat treatment and with the polymer treatment is evident, and in agreement with the marked reduction in the open porosity values.

Crushing Strength of Aggregates. The crushing strength values of the aggregates are presented in Table 4 alongside with porosity values and their relative change with reference to the untreated Lytag aggregate. The crushing strength value for the L1300RC aggregate is extremely low and this could be accounted for by numerous cracks observed on its surface, reflecting probably the influence of thermal gradients which were the biggest in this case. The other aggregates did not seem to be damaged and the differences in their crushing strength values could therefore be discussed in terms of the porosity and the nature of the solid which comprises the aggregates.

The differences in crushing strength could not be correlated only with differences in porosity. For example, aggregates L1250 and 1300RC have lower porosity than the original Lytag aggregate, but their strength is lower. This may be due to several influences: change in the mineralogical composition and cracks and internal defects due to thermal stresses. The much lower strength of the L1300 RC aggregate could be clearly related to the renewed stresses, as cracking could be clearly observed.

Referring to Table 4 all the aggregates can be classified into three groups according to their strength:

i) L1250, L1300SC aggregates with crushing strength similar to the original Lytag aggregate (crushing strength of about 15MPa).

TABLE 4

Physical and Mechanical Properties of the Aggregates Properties, Relative to Untreated Original Lytag, %, Due to Thermal Treatments

Aggregate type	Crushing Strength MPa	Porosity, % vol.		Properties relative to untreated Lytag, %		
		Total	Open	porosity change total	open	crushing strength change
Lytag	14.9	37.3	19.1	100	100	100
L1200	19.3	36.2	14.1	97.3	73.8	129.5
L1250	14.7	27.9	12.4	75.0	65.0	98.7
L1300RC	6.0	33.4	7.1	89.8	37.2	40.4
L1300SC	15.6	32.6	10.6	87.6	55.5	104.7
polymer	19.9	30.6	8.4	82.3	44.0	133.6

TABLE 5

Compressive Strength of Lightweight Aggregate Concrete

Group	Aggregate Type	Aggregate Crushing Strength, MPa	Concrete compressive strength, MPa at:				
			1 day	7 days	28 days	56 days	90 days
I	Lytag	14.9	10.5	38.8	45.4	53.6	57.7
	L1250	14.7	9.3	34.6	41.0	52.8	57.5
	L1300SC	15.6	8.2	34.5	40.5	54.5	58.5
II	L1200	19.3	12.1	46.9	54.8	57.8	60.9
	polymer	19.9	9.8	35.7	43.6	45.0	45.3

ii) L1200 and polymer treated aggregates with similar crushing strength (~19.5 MPa) but greater than the original Lytag.

iii) L1300RC with the smallest crushing strength.

Strength of Concretes. The values of compressive strength of concretes prepared with the same effective w/c ratio of 0.40 (after allowing for the absorption of the aggregate) are shown in Table 5. The data is grouped into two classes of concretes each made of aggregates of similar crushing strength. It can be seen that comparison within each class shows that the strength values are not necessarily the same even though the aggregates and the matrix are of the same quality (Fig. 8).

For example, concretes with L1200 and polymer treated aggregates are quite different in their strength at all ages in spite of the fact that the two aggregates are of similar strength (Fig. 8b). When comparing between the different groups, it can be seen that higher strength aggregate does not necessarily lead to higher, or proportionally higher strength concrete. For

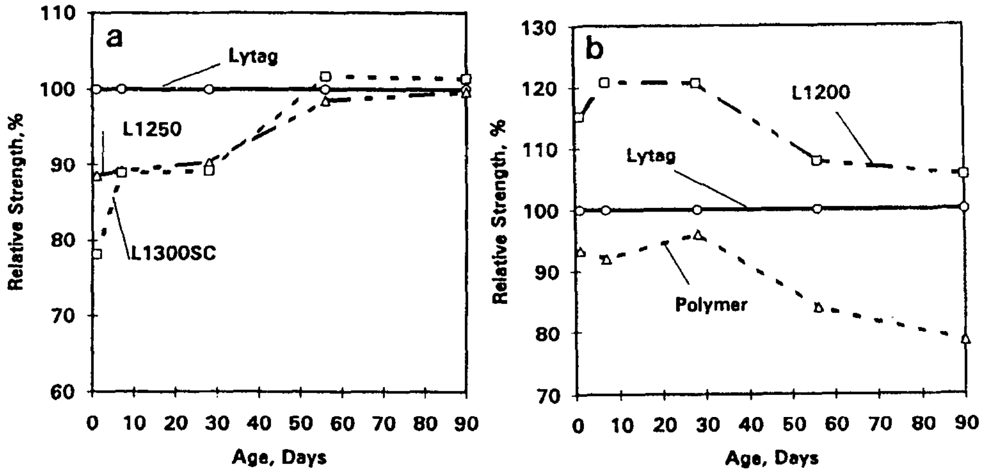

FIG. 8.

Strength of concretes relative to the concrete with untreated Lytag aggregate: (a) concretes of group I, (b) concretes of group II.

example the L1200 aggregate which is much stronger than the untreated Lytag aggregate (~30% stronger) yields at the age of 90 days a concrete which is only ~5% stronger (Fig. 8b); the strength of the polymer treated aggregate concrete is significantly less (20-30%) than the untreated Lytag aggregate concrete (Fig. 8b) although the untreated Lytag aggregate is considerably weaker.

Therefore there is reason to suggest that the mechanical properties of lightweight aggregate concrete can not be predicted accurately only on the basis of the strength of the aggregate and the effective w/c ratio (i.e. strength) of the paste matrix. Other parameters should be considered: (i) It can be seen in both groups that at early age lower strength was obtained with aggregates of smaller absorption; (ii) In group I (Fig. 8a) the closure of the gap in strength at later age between the concrete from untreated aggregate and the heat treated aggregates might be attributed to the higher pozzolanic activity of the latter (Fig. 2). To resolve the influence of these factors and their origin, additional work to study the internal structure of these concretes is needed, and this will be the subject of a future publication.

Conclusions

1. All the treatments led to a decrease of in the total and open porosity and in the water absorption.
2. The pozzolanic activity of the outer shell of the aggregate increased with an increase in the treatment temperature. This is suggested to be the result of the increase in either the content or activity of the glassy phase.
3. The reduction in the porosity of the aggregate was not always accompanied by an increase in the crushing strength. This may be due to additional influences associated with mineralogical changes and internal thermal stresses.
4. The treatment with polymer resulted in impregnation which led to reduction in porosity and absorption of the aggregate and increase in its strength.
5. The strength of the concretes prepared with the different lightweight aggregates did not necessarily correlate with the strength of the aggregates although the matrix was always of the same effective w/c ratio. The matrix w/c ratio.

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