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Interfacial Interactions in Lightweight Aggregate Concretes and their Influence on the Concrete Strength

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Abstract

The interactions between sintered fly ash lightweight aggregates and the matrix in portland cement concretes was studied to resolve factors other than aggregate strength which influence the concrete strength. Aggregates of variable properties were produced and concretes of equal effective water/cement ratio were prepared and tested for strength and microstructure. It was found that differences in concrete strength could not always be accounted for by differences in the aggregate strength. These trends could be related to physical and chemical interfacial processes, which have an influence on the overall strength beyond that of the aggregate strength. The physical process identified was densification of the interfacial transition zone due to absorption of the aggregates; this process has considerable influence already at early age. The chemical processes were associated with pozzolanic activity of the aggregate and deposition of CH in the pores in the shell of the aggregate; these processes became effective only at later age, beyond 28 days. The enhancement in strength due to these influences ranged between 20 and 40%. Such influences should be taken into account when predicting the concrete strength or in the design of lightweight aggregate of optimal properties.

Keywords: Aggregates, concrete strength, fly ash.

INTRODUCTION

There is considerable increase in the application of high strength lightweight concretes using lightweight aggregates. Therefore, there is renewed interest in the role of aggregates in such concretes, particularly since they are the weak link in these systems. Usually, the strength of such concretes is believed to be controlled by the strength of the aggregates and the strength of the paste. Recent studies into the microstructure of such concretes suggest that the interaction of the lightweight aggregate and the matrix can be quite different than that of normal aggregate.[1–8] The interfacial microstructure can be quite similar to that of normal concrete if the aggregates are wetted, but it can be considerably different and much denser if the aggregate is used dry. In some studies it has been suggested that pozzolanic reaction between the aggregate and the paste matrix is a factor contributing to strength[4,5] while in others no such interaction was observed.[3,7]

In view of the influence that the lightweight aggregate–paste matrix interaction may have on the concrete strength, a systematic study was undertaken with sintered fly ash lightweight aggregates to resolve some of the mechanisms by which they interact with the matrix, and assess their potential influence on the strength of the concrete. To achieve this goal, commercial Lytag aggregate was modified by heat and polymer treatments to obtain aggregates with variable properties, i.e. differences in strength, absorption and pozzolanic activity. The inter-

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action of these aggregates with the paste matrix was studied. In a previous paper[9] the properties and microstructure of the aggregates as well as the strength of concretes of equal effective water/cement ratio were reported. It was shown that the differences in strength could not be accounted only on the basis of the aggregate strength. In the present paper the microstructural study of these concretes is reported, in particular the interfacial characteristics. The interfacial processes resolved can explain differences in the strength of lightweight aggregate concretes which could not be accounted for merely by the strength of the aggregate.

EXPERIMENTAL

The lightweight aggregates used in the present study were sintered fly ash. Commercial Lytag (UK) aggregate was treated by heat (heating to 1200, 1250 and 1300°C and rapid or slow cooling afterwards) or by polymer (partial impregnation and hydrophobization) to obtain aggregates with controlled difference in absorption and pozzolanic reactivity. Details of the aggregate preparation and their internal structure are given in Ref. 9. Table 1 summarizes their main characteristics. It can be seen that they can be classified according to their strength into two groups with each having aggregates of similar strength but different in other properties. Accordingly, the concretes were also classified into two groups.

Concretes were prepared with aggregate blend of 51% lightweight aggregate and 49% graded normal weight fine aggregate, at an effective w/c ratio of 0·40 (after allowing for the early absorption of the aggregate). Details of concrete compositions and preparation methods and their strength values are given in Ref. 9. They were all demolded after one day and than cured continuously in water at 20°C up to 90 days. In the present study additional concretes were prepared also using normal aggregate, and curing of the lightweight and normal aggregate concretes at elevated temperature to resolve pozzolanic effects. This curing procedure consisted of 28 days in water at 20°C followed by water curing at 60°C up to a total curing time of 90 days.

The concretes were tested periodically for compressive strength (70 mm cubes), and their internal structure was characterized by SEM observations and micro-chemical analysis of polished surfaces. The specimens were impregnated with low viscosity epoxy and polished to obtain a flat surface for micro-analysis. Energy dispersive X-ray analysis was used to determine the Ca and Si contents, and profiles of C/S ratio across the aggregate–paste interface were drawn.

RESULTS AND DISCUSSION

SEM observations

SEM observations were carried out at one day and at a later age of 90 days. At one day there seemed to be intimate contact between the aggregate and the matrix only in the concrete from the original Lytag aggregate (Fig. 1(a)). In all the treated aggregates separation was observed, as shown for two examples in Fig. 1(b) and (c). The separation does not necessarily mean that there is a void in the interfacial zone; it could reflect the presence of a very porous material which shrinks considerably during the drying in the preparation of the specimens for the SEM observation, with eventual separation. Indeed, observations at the

Table 1. Properties of lightweight aggregates

Group	*Name*	*Treatment*	*Crushing strength, MPa*	*Water absorption, %Vol.*	*90 Days pozzolanic activity of the outer shell**
	Lytag	untreated	14·9	14·5	0·15
I	L1250	1250°C, rapid cooling	14·7	9·7	0·40
	L1300SC	1300°C, slow cooling	15·6	9·2	0·52
II	L1200	1200°C, rapid cooling	19·3	10·6	0·28
	polymer	polymer	19·9	3·8	—

*Determined on the basis of reaction with CH.

paste surface exposed after the aggregate removal show a denser microstructure in the concrete with the untreated Lytag aggregate (Fig. 2(a)) and much more porous material in the treated aggregates as seen in the two examples shown in Fig. 2(b) and (c). It should be noted that the differences are not only in the overall porous nature but also in the type of microstructure detected, showing more needle-like material, apparently ettringite, in Fig. 2(b).

At 90 days there seems to be intimate contact at the interface in all of the concretes, except the one with the polymer treated aggregate, which even at 90 days showed separation at the

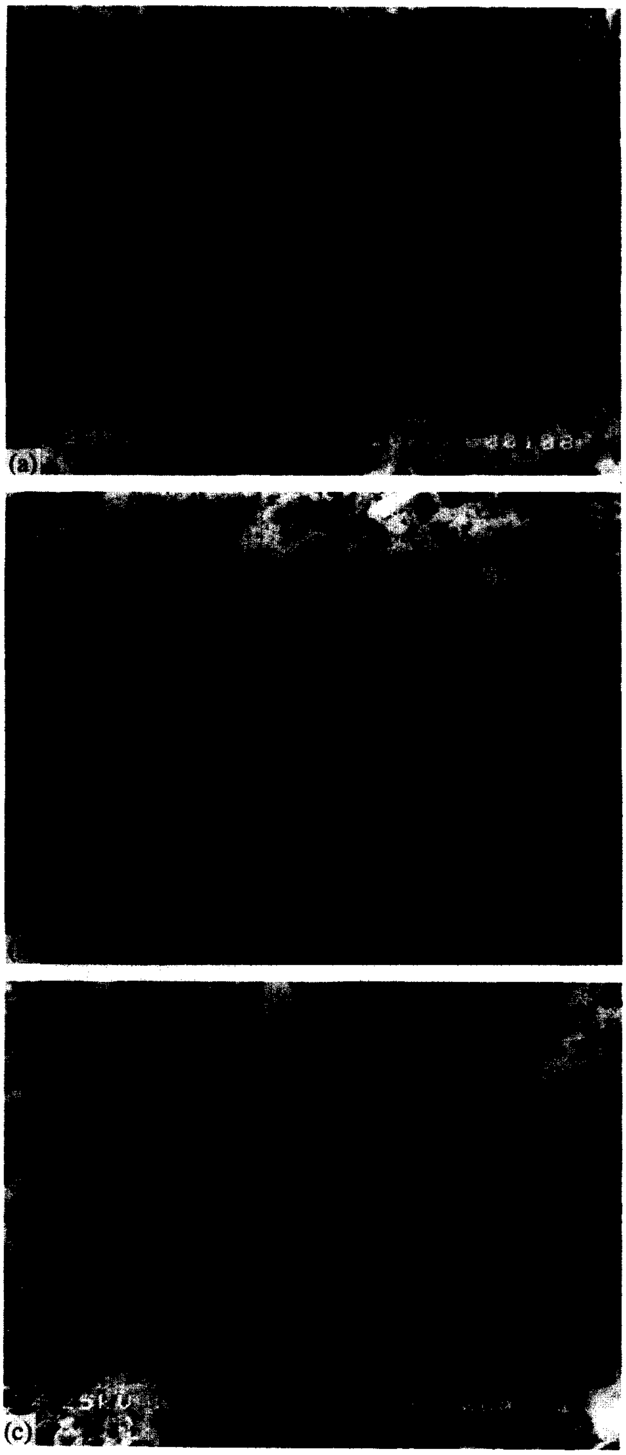

Fig. 1. The aggregate-matrix interface at one day: (a) untreated Lytag, (b) L1250 and (c) polymer treated.

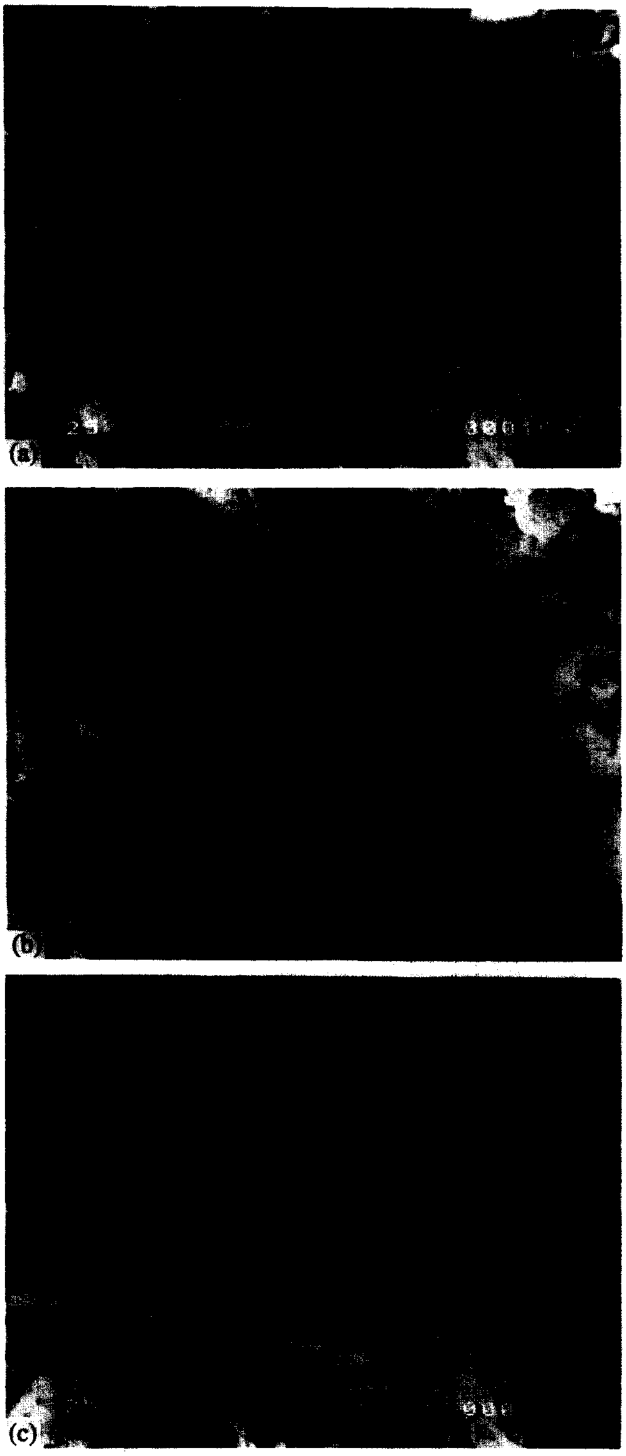

Fig. 2. View of the matrix side of the aggregate-matrix interface: (a) untreated Lytag, (b) L1300 and (c) polymer treated.

interface (Fig. 3). The surface of the original Lytag and heat treated aggregates is quite fuzzy and the structure of the aggregate is masked without being able to resolve its porous nature (Fig. 4(a) and (b)). It seems that the pores in the aggregates have been penetrated by hydration products, and in the case of the untreated Lytag aggregate there is evidence for presence of large crystals in the pores (Fig. 5) whereas in the treated aggregates no clear crystalline morphology could be observed. In contrast to these aggregates, the polymer treated aggregate preserved its original nature, and remnants of fused fly ash particles and some pores could be clearly observed (Fig. 4(c)).

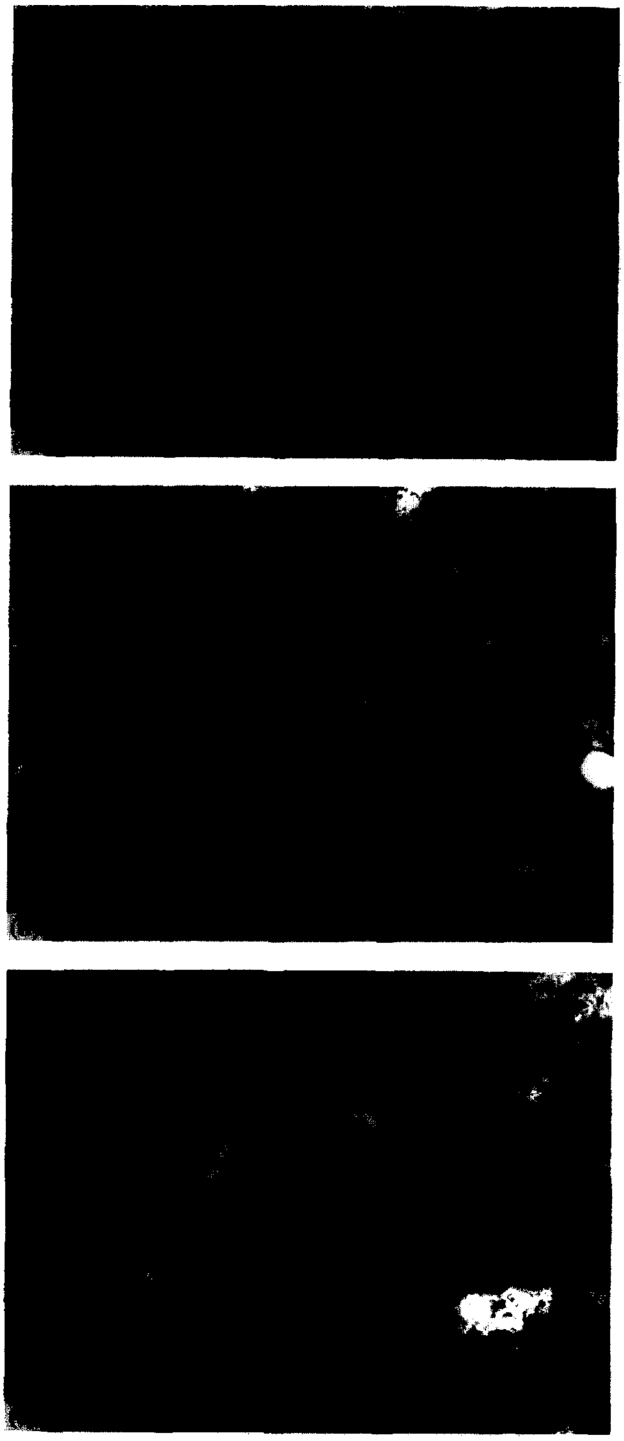

Fig. 3. The aggregate–matrix interface at 90 days: (a) untreated Lytag, (b) 1300SC and (c) polymer treated.

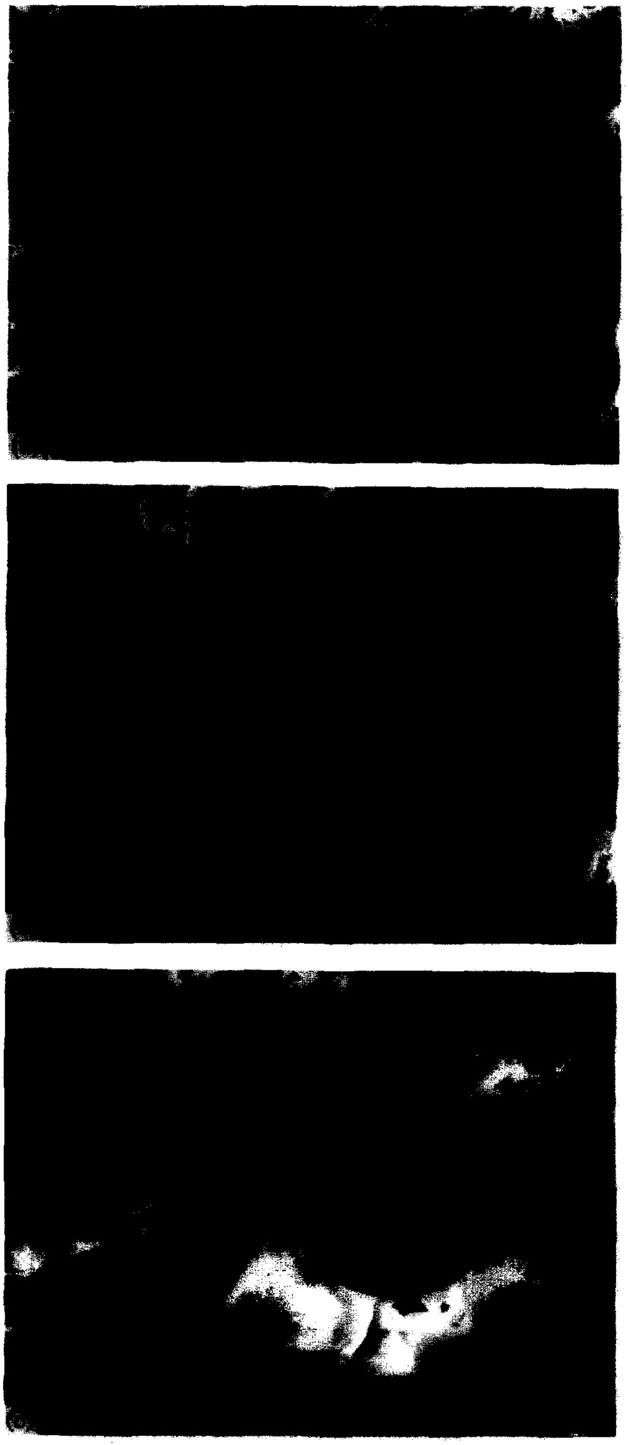

Fig. 4. View of the aggregate side of the aggregate–matrix interface at 90 days: (a) untreated Lytag, (b) L1300SC and (c) polymer treated.

Fig. 5. High magnification of the aggregate surface at the aggregate–matrix interface in 90 days cured concrete with untreated Lytag aggregate.

Composition of the interfacial transition zone

The 1 day and 90 day compositions across the interface from the paste into the aggregates are provided in Figs 6–8, in terms of C/S ratio profiles. The C/S ratio of the aggregate shell itself is about 0·15 and that of the bulk paste is about 3. It can be seen in Figs 6–8 that in the vicinity of the actual interface there are deviations from these bulk values which extend into the paste matrix as well as into the aggregate.

At one day the C/S ratio curves drop sharply just at the interface to the values typical of the aggregate shell (~0·15). This is characteristic of all the three aggregates in Figs 6–8. However, in front of the interface, towards the paste matrix, there is a difference between the untreated Lytag (Fig. 6) and the heat treated one (Figs 7 and 8). In the former the C/S curve is

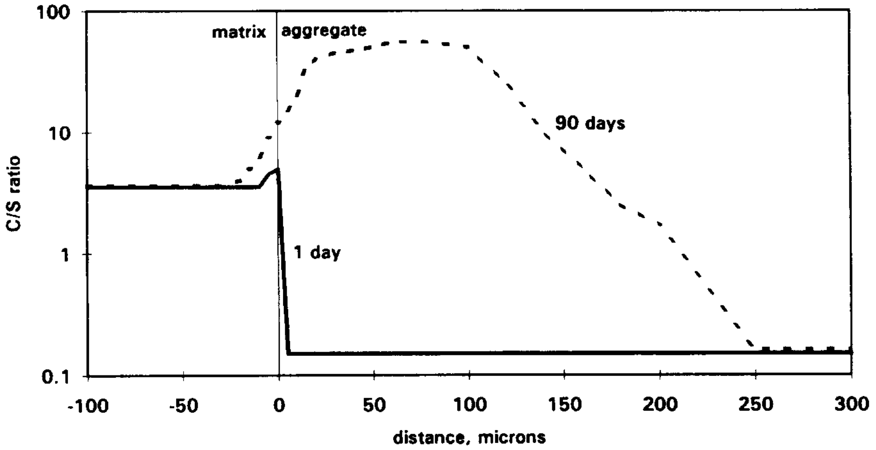

Fig. 6. C/S ratio profiles across the aggregate–matrix interface in 1 and 90-day-old concretes with untreated Lytag aggregate.

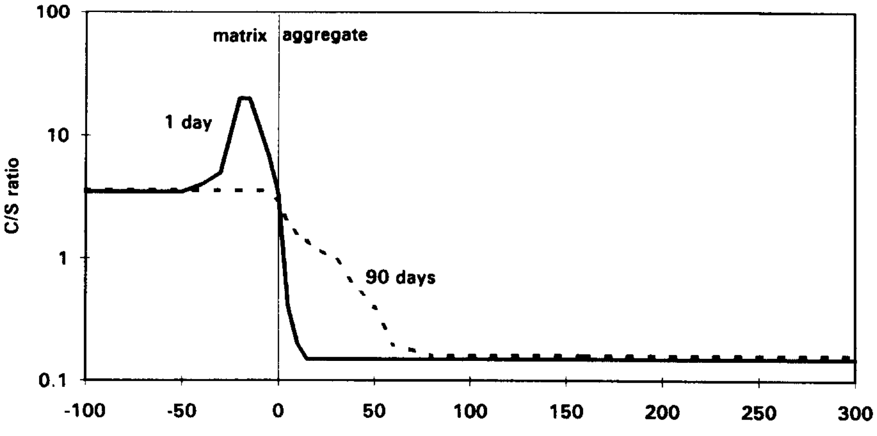

Fig. 7. C/S ratio profiles across the aggregate–matrix interface in 1 and 90-day-old concretes with L1250 aggregate.

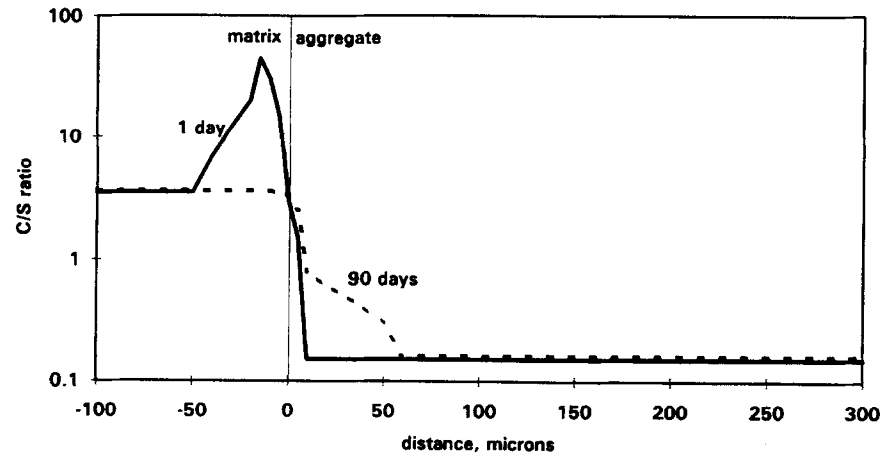

Fig. 8. C/S ratio profiles across the aggregate–matrix interface in 1 and 90-day-old concretes with L1300SC aggregate.

almost horizontal at a value of ~3·5, from the bulk paste up to the actual interface. In the latter there is a zone in front of the actual interface towards the bulk matrix where the C/S ratio is high, about 10–40, and it extends over a distance of several tens of microns. This trend is also characteristic of the two other treated aggregates, at 1200°C and the polymer treated one (not shown here). The width of the interfacial transition zone in front of the aggregate can be estimated as the zone in which the C/S ratio is high, and it is about 4, 25 and 40 μm for the original Lytag aggregate and the 1200 and 1300°C treated ones, respectively. The lower value for the untreated aggregate may be accounted for by the higher absorption which prevents accumulation of water at the interface; accumulation of such water in the fresh concrete is believed to be the cause for the formation of the porous zone in the vicinity of the aggregate surface, resulting in a more porous microstructure which is rich in CH. Both of these characteristics were observed here, in the SEM micrographs and in the composition profiles. A plot of the width of the interfacial transition zone as a function of the 6 h water absorption of the aggregates (Fig. 9) shows a clear relation, with the width of the interfacial transition zone decreasing with the increase in the absorption, as might be expected on the basis of the explanation provided here.

This trend changes at 90 days, where a high C/S ratio curve seems to be 'penetrating' deep into the aggregate in the untreated Lytag aggregate (Fig. 6), whereas in the heat treated aggregates, at 1250 and 1300°C, the C/S ratio curve is declining gradually within the aggregate boundaries, from values of about 3 at the interface to the characteristic aggregate C/S ratio of about 0·15. This occurs at a distance of about several tens of microns into the aggregate (Figs 7 and 8). These results suggest that in the original Lytag aggregate the pore solution can penetrate effectively into the pores of the aggregate, due probably to its large absorption capacity, and deposits of CH are formed in the aggregate pores, as evidenced by the high C/S ratio and the SEM observations. In the higher temperature treated aggregates there is probably absorption of this kind, although to a smaller extent, and since the aggregates are more pozzolanic, CSH rather than CH is formed preferentially. In these aggregates, the

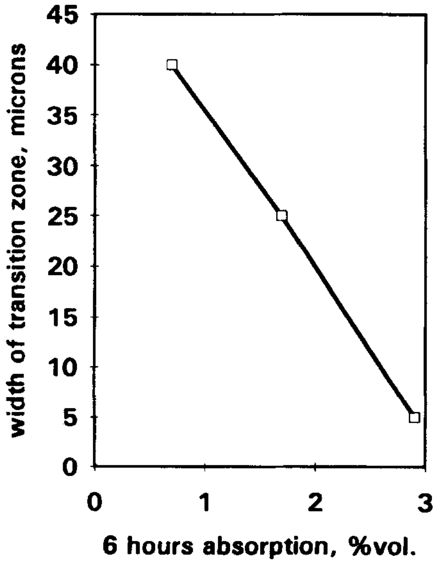

Fig. 9. Relation between interfacial transition zone width at one day and the 6 h absorption of the lightweight aggregate.

reduction in C/S ratio in the interfacial transition zone in front of the aggregates from high values of up to 20 at 1 day, to about 3 at 90 days suggest that the larger deposits of CH at early age were the source for the Ca ions that penetrated into the aggregate, to become involved in the pozzolanic reaction.

Strength

The effect of the aggregate type on the strength levels at 1, 28 and 90 days is shown in Fig. 10.

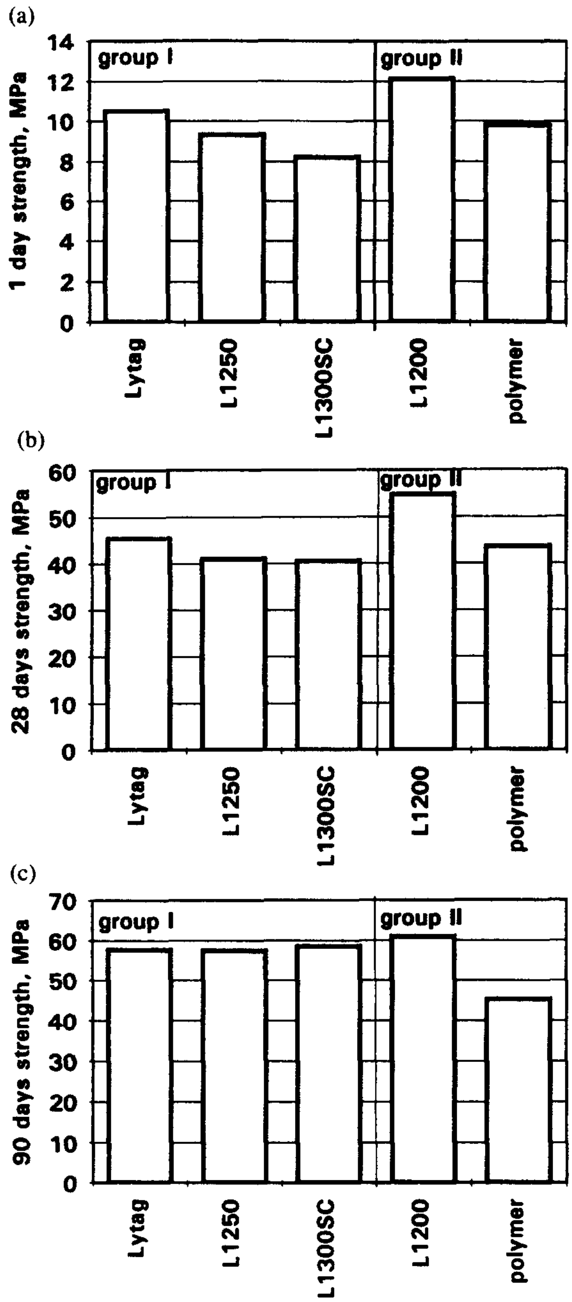

Fig. 10. Compressive strength values at 1, 28 and 90 days of concretes of groups I and II.

Within each group of equal aggregate crushing strength (groups I and II) the concrete strength values are not equal. In group II the polymer treated aggregate is consistently weaker by about 20% than the heat treated aggregate. In group I the trends change with time: at early age the strength of the concrete is smaller for the higher temperature treated aggregate, but with time the differences diminish, and by 90 days they are all practically equal. When comparing between the groups it can be seen that the higher strength heat treated aggregate in group II provides a concrete which at early age is stronger than all the concretes in group I which is in agreement with the higher strength of the L1200 aggregate in group II. However, this difference diminishes with time and by 90 days the concretes of group I are practically of the same strength as the L1200 aggregate concrete in group II, inspite of the higher crushing strength of the latter aggregate. The polymer treated aggregate in group II always gave a weaker concrete than those in group I, inspite of its higher strength compared to all of the aggregates in group I.

The fact that the trends in strength observed at one day change over time, suggests that there is a 'dynamic process' whose nature can be better resolved when plotting for each of the systems the strength values relative to the 28 days strength (Fig. 11). This figure contains also, for comparison, the relative curve for normal aggregate concrete of the same w/c ratio. Up to 28 days the relative strength values

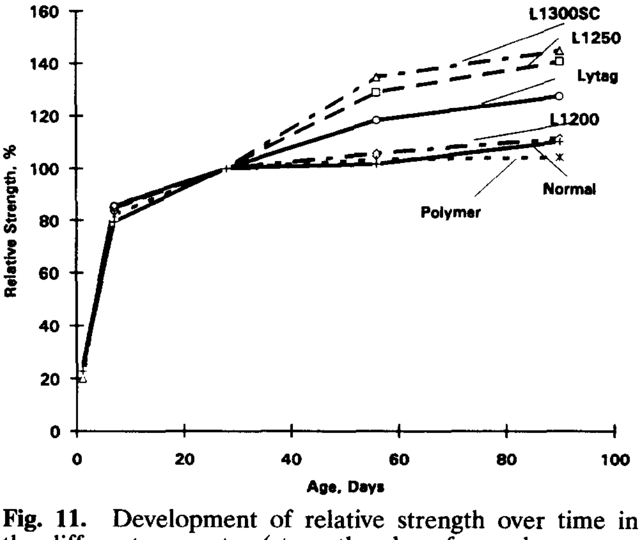

Fig. 11. Development of relative strength over time in the different concretes (strength values for each concrete relative to its 28 days strength).

are similar in all of the concretes. Beyond that they can be classified into three types: (a) small increase in strength, less than 5% at 90 days (characteristic of the normal aggregate concrete and the aggregates of group I — polymer treated and 1200°C treated), (b) mild increase in strength, about 20% at 90 days (characteristic of the untreated Lytag aggregate concrete), and (c) marked increase in strength, of about 40% at 90 days (characteristic of the concretes with aggregates treated at 1250°C and 1300°C).

The differences in the trends at early age and later ages suggest that different mechanisms control the strength values at these two ages and they are superimposed on the effects of the aggregate strength. When comparing the differences at early age in concretes prepared from aggregates of similar strength it seems that the lower strength concretes are the ones with aggregates of smaller absorption capacity, as can be seen from the relations in Fig. 12. This trend can be explained by the formation of a more porous interfacial transition zone with the lower absorption aggregates (Fig. 9). This also shows up in the SEM observations indicating aggregate–matrix separation in the lower absorption aggregates (Fig. 1).

At later ages, the SEM observations indicate a very dense transition zone in all of the concretes except the one of the polymer treated aggregate (Figs 3 and 4). This may account for the much lower strength values of the concrete with the polymer treated aggregate which is maintained even at this age. In all other concretes the interfacial transition zone seems much denser than in normal strength aggregates and it is difficult to resolve the border between the aggregates and the matrix (Figs 3 and 5), in contrast to normal aggregate concrete where deposits of CH and some porosity can be observed at the interfacial transition zone.[10] Although the interfacial region was dense in all the heat treated and untreated Lytag aggregates, differences in the composition of this zone were seen. In the L1250 and L1300SC aggregates, the C/S ratio curves suggest the presence of CSH within a zone of about 50 μm within the outer shell of the high temperature treated aggregate, which could be correlated with their higher pozzolanic activity (Table 1). This can account for the relatively high increase in the strength of the concretes prepared from these aggregates after 28 days (Fig. 11). It more than compensates for their lower initial strength due probably to some reinforcement of the outer shell of the aggregate and bond enhancement due to the continuity in the CSH outside and inside the aggregate. In the concretes prepared with the lower temperature heat treated aggregate (1200°C) and the normal aggregate no pozzolanic activity was observed and no significant strength increase occurred beyond 28 days. The concrete from the untreated Lytag aggregate did not seem to behave according to these trends, and although it was not pozzolanic it showed some strength enhancement beyond 28 days. This might be explained by the formation of CH deposits in the shell of this aggregate which probably strengthens it, but are probably less effective in enhancing the bond as suggested to occur in the pozzolanic active aggregates.

In order to check the hypothesis of the influence of pozzolanic activity of the aggregates, three concretes were cured beyond 28 days in 60°C water: concretes with normal aggregate and the original Lytag, both of which are non pozzolanic, and the concrete with the 1300°C aggregate (1300SC) which had the highest pozzolanic activity. The results presented in Fig. 13 show, as expected, that the high temperature curing did not affect the first two concretes. However it enhanced the strength of the third concrete, which could be attributed to mobilizing to a greater extent its pozzolanic activity.

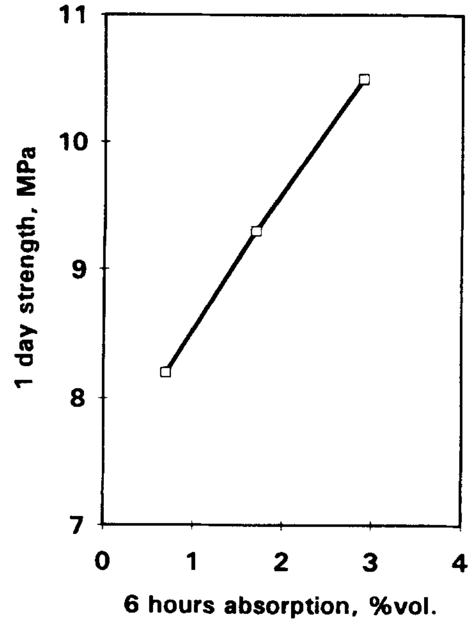

Fig. 12. Relation between the one day compressive strength in group I concretes and their 6 h water absorption.

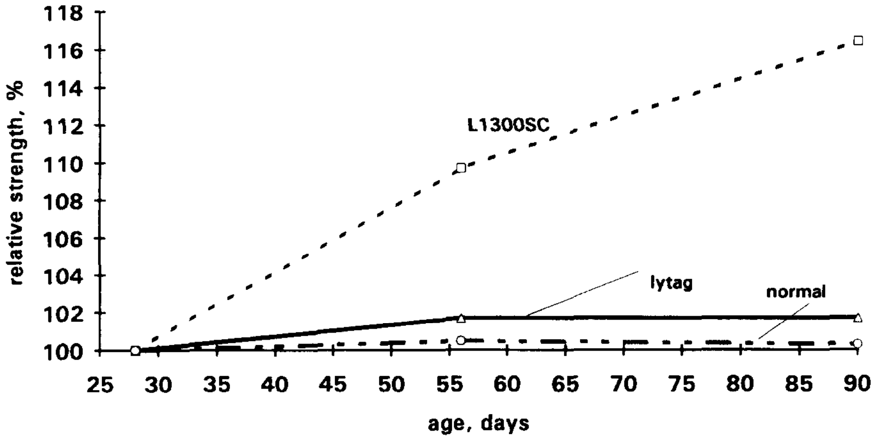

Fig. 13. Relative strength curves of concretes cured at 60°C beyond 28 days (strength values at 60°C relative to 20°C curing).

CONCLUSIONS

(1) Lightweight aggregates of similar strength do not necessarily yield concretes of equal strength even if the matrix is of the same effective w/c ratio. Thus, factors in addition to the aggregate strength should be considered.

(2) The additional influences that should be considered were found to be associated with the physical and chemical characteristics of the aggregates, both of them affecting the overall strength by processes which take place at the interfacial transition zone. In the aggregates this zone extended also into the aggregate itself.

(3) The physical process occurs at early age and is governed by the absorption of water into the aggregate. Higher absorption eliminates accumulation of water in the fresh matrix in the vicinity of the aggregate. As a result the interfacial transition zone in lightweight aggregates of higher absorption is denser. Thus, for lightweight aggregates of equal strength, the aggregate of higher absorption will provide higher strength concrete due to its denser interfacial transition zone.

(4) The chemical process occurs at later age. Two types of processes were resolved here: pozzolanic reaction between the aggregate and the alkaline pore solution which penetrates into it, and an 'impregnation' process in which CH deposits in the pores of the aggregates. The latter is more likely to occur in aggregate having higher absorption and bigger pores.

(5) The contribution of the processes identified in enhancing the concrete strength can be evaluated by comparison of the relative strength values of the different concretes:

- Physical processes: At early age the strength difference in concretes having aggregates of similar strength is about 25% due to differences in 6 h absorption capacity of about 200%; the higher strength is in the concretes with higher absorption aggregates.
- 'Impregnation' mechanism can result in strength increase of about 20% at 90 days (relative to 28 days).
- Pozzolanic activity can lead to strength increase of about 20% at 90 days (relative to 28 days).

(6) This is a basis for considering these mechanisms for controlled production of lightweight aggregates intended for use in high strength lightweight concrete.

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