The microstructure and ageing of cellulose fibre reinforced autoclaved cement composites

A. Bentur* and S. A. S. Akers†

Synopsis The correlations between microstructural changes after ageing and the mechanical performance of autoclaved cellulose-cement composites were studied. The autoclaved composite did not undergo marked microstructural changes during ageing, and this can account for the relatively small changes in mechanical property development during exposure to natural weathering and accelerated ageing tests. In particular autoclaved cellulose cement products indicated significantly less 'petrification' of the cellulose fibres with age when compared with normal cured products of similar mix composition [1].

Keywords Fibre cement composites, microstructure, cellulose fibres, ageing, scanning electron microscopy, autoclaving, testing, composite materials.

INTRODUCTION

The present paper deals with a study of the durability and ageing of autoclaved cellulose fibre reinforced cement composites. In previous work [1] the ageing processes for room temperature cured composites were investigated. In this composite, natural ageing resulted in an increase in flexural strength and E-modulus [2]. These changes in properties could be simulated by an accelerated test in which the composite was exposed to wetting and drying cycles in CO_2 rich environment. It was suggested that ageing in a CO_2 rich environment and natural weathering resulted in densification of the matrix around the fibres, and the petrification of the fibres, thus leading to increase in strength and E-modulus of the product. The petrification of the fibre was associated with build up of reaction products within the core of the cellulose fibre, and perhaps also within the fibre cell wall, both of which led to effective strengthening and stiffening of the fibre.

The effect of autoclaving on the properties of the composites before and after ageing has been reported by Akers et al. [2], and some of this data is reproduced here in Table 1. The results indicate that for autoclaved products the changes in mechanical properties during ageing were less significant when compared with a normal cured product.

The objective of this paper is to present a plausible explanation describing the chemical microstructural changes in an autoclaved product which may be related to variations in the mechanical properties of the products during ageing.

EXPERIMENTAL DATA

The experimental work in the present study deals mainly with evaluation of the structure of the composites before and after ageing, using scanning electron microscopy for microstructural characterisation of the interface and thermal gravimetric (TG) and X-ray diffraction for determination of the bulk phase composition. The details of these experimental procedures are provided in reference [1] and description of specimen preparation, composition and testing for mechanical properties are provided in reference [2].

RESULTS

The XRD and TG data indicate considerable carbonation in the composites exposed to natural ageing and accelerated ageing in a CO_2 rich environment (Table 2, Figure 1). The extent of carbonation from data of this kind is based on the evaluation of the increase in CC* content.

* Building Research Station, Department of Civil Engineering, Technion, Israel Institute of Technology, Haifa, Israel.
† Ametex AG, Research and Development, Niederurnen, Switzerland.

Received 15 November 1988 Accepted 16 January 1989

0262-5075/89/11205111/$02.00

* Cement chemistry notation is used: $C = CaO$, $S = SiO_2$, $A = Al_2O_3$, $F = Fe_2O_3$, $H = H_2O$, $\bar{S} = SO_3$

Table 1 Mechanical properties of composites before and after ageing (2)

	Properties of composite			
	Modulus of rupture (N/mm^2)		Degree of polymerisation of the ceullulose fibres	
Composite type	Unaged	Aged	Unaged	Aged
Normal curing	16.4	23.9* 21.9†	581	491* 435†
Autoclaved	23.0	27.2* 21.8†	703	475* 504†

* Natural ageing, 4 years
† Accelerated ageing in a CO_2 rich environment, 3 months

It is interesting to note, that in the autoclaved composite there is obviously no CH in the unaged material and therefore the marked increase in the $C\bar{C}$ content during ageing could apparently be the result of carbonation of the calcium silicate hydrate.

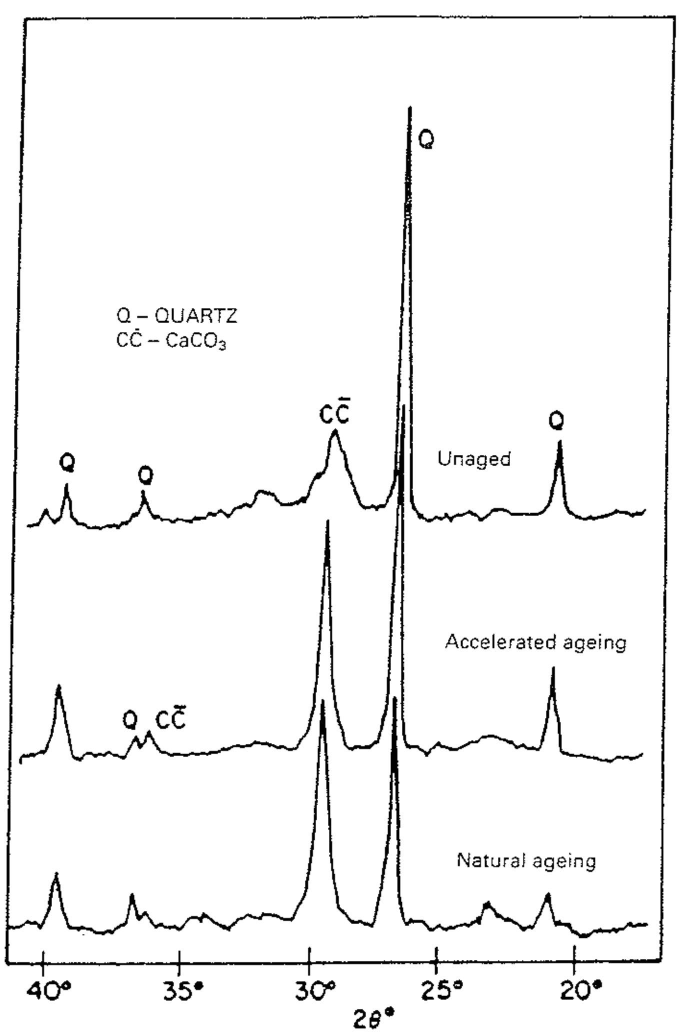


Figure 1 XRD patterns of autoclaved composites before and after ageing

The degree of polymerisation of the fibres (see Table 1) appeared to reduce with ageing, although at a relatively mild rate. This is not accompanied by any significant change in the mechanical properties of the composites.

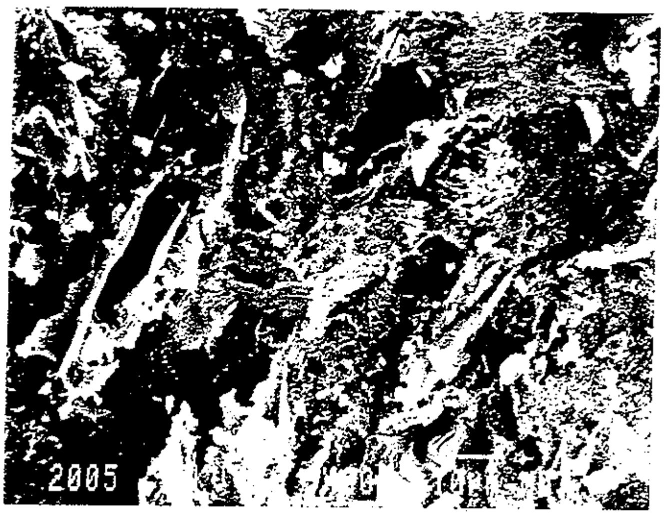
Table 2 $C\bar{C}$ content in autoclaved composite before and after ageing

	$C\bar{C}$ content, % Wt
Unaged	0
Accelerated ageing (CO_2 environment 3 months)	7.9
Natural ageing (4 years)	12.9

SEM observations of the interface (unaged and aged products)

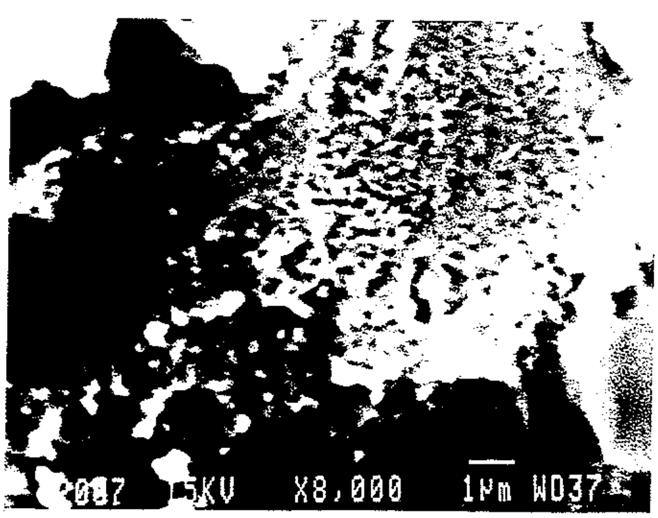
Generally the typical mode of fibre failure in this composite was a brittle one, with fibres breaking at the fractured surface (Figure 2a,b). Only seldom was fibre pull-out seen (Figure 3). The matrix at the interface was usually denser (Figure 2b,c and Figure 4) than observed in the unaged normally cured composite, although around some fibres a mixed mode of dense and more porous matrix could be seen (Figure 2b). The dense matrix was able to form an intimate contact with the fibre at the interface (Figure 4).

The mode of failure and interfacial microstructure of the accelerated and natural aged composites were similar, and therefore will be described together. The mode of fracture and interfacial microstructure were similar to those observed in the unaged composite. Brittle fibre fracture was dominant with the matrix around the fibres being dense (Figure 5). In contrast to the unaged composite, a gap could be frequently seen between the matrix and the fibre. In the accelerated aged composite there was also some indication of pull-out in a few fibres to a limited extent (see for example Figure 6) or more extensively (Figure 7). In all these instances the



(a)

(b)



(c)

Figure 2 (a) Brittle fracture in an unaged, autoclaved composite, general view; (b) high magnification of the left fibre in (a); (c) high magnification of the dense matrix to the right of the fibre in (b)

Figure 3 Fibre pull-out in unaged autoclaved composite

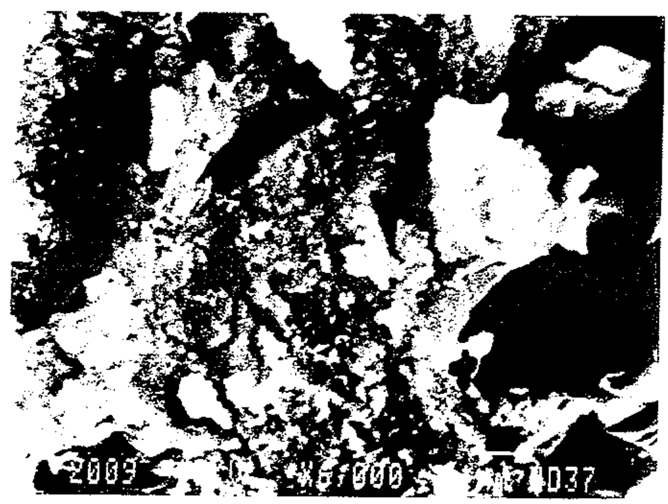


Figure 4 Dense matrix and the intimate contact it forms with a fibre (seen at the right side) in an unaged autoclaved composite

matrix was quite dense around the fibres which in turn fractured in a brittle manner. The fibres also seemed to twist during pull-out.

DISCUSSION

In the unaged composite, the matrix in the vicinity of fibres was relatively dense. The common mode of failure was brittle hollow. This is quite different from the observations of the unaged normal cured composite reported earlier [1], which indicated the presence of a very open microstructure in the vicinity of the fibres, leading to a pull-out mode of failure. The dense matrix and better fibre/matrix bond in the autoclaved composite can lead to increased E-modulus of the product with age. During ageing, no major microstructural changes had taken place, neither in the fibres nor in the matrix surrounding them. The common mode of failure

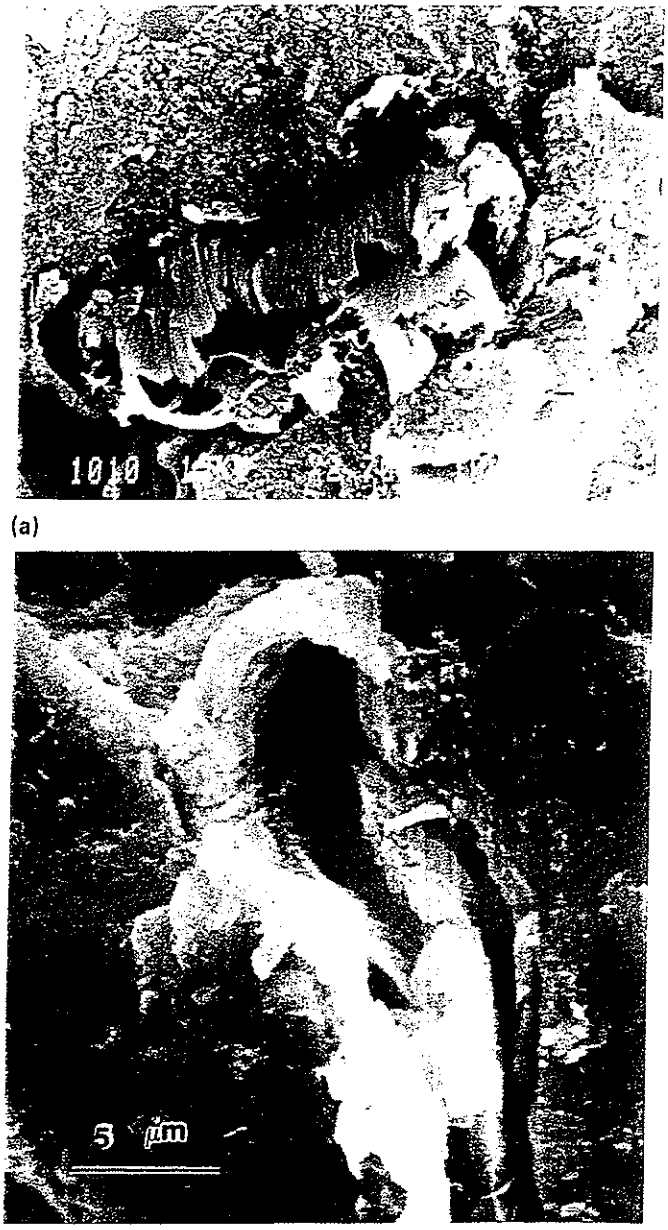


Figure 5 (a) Brittle failure in a naturally aged autoclaved cellulose composite; (b) a second example of this type of failure showing in (a)

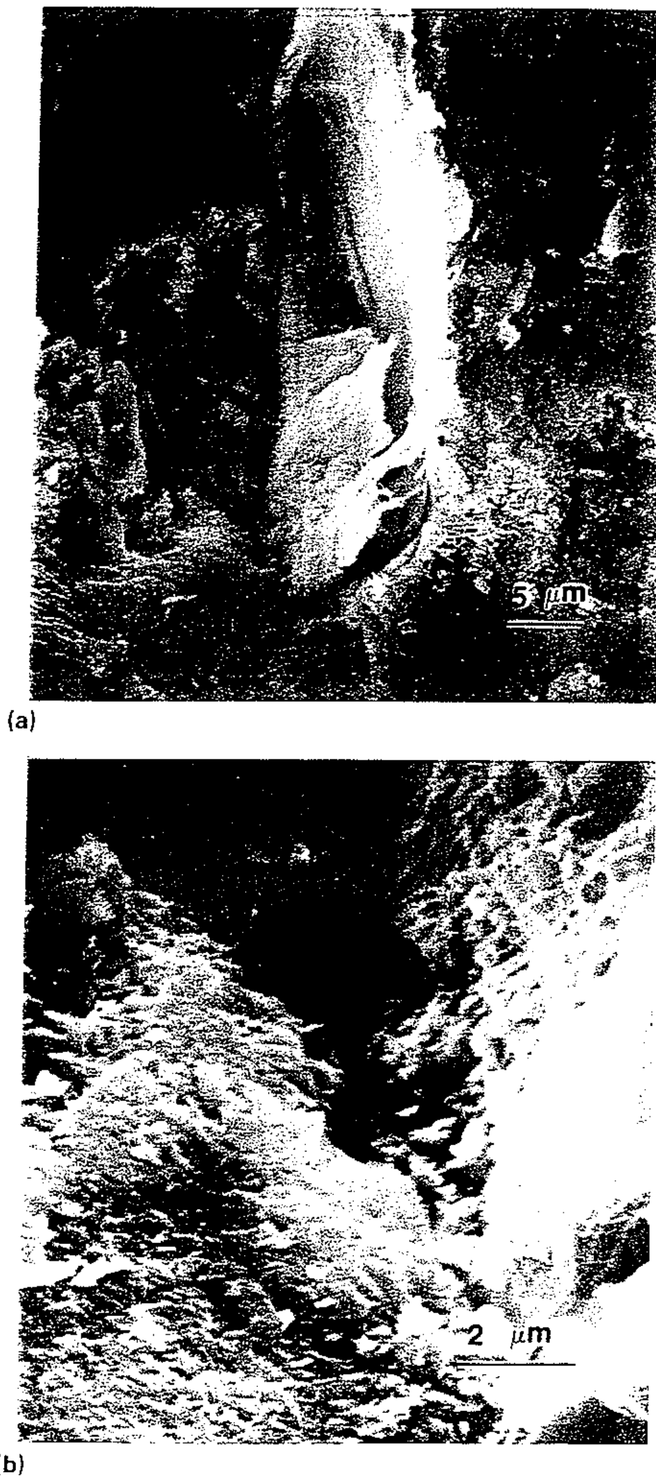


Figure 6 (a) Brittle failure in an accelerated aged autoclaved composite showing a small extent of fibre pull-out; (b) high magnification around the bottom left of the fibre in (a) showing the dense matrix

remained 'brittle-hollow' [1]. Some small spaces could be seen to have opened between the fibre and matrix, due possibly to shrinkage of the matrix and fibres, induced during the wetting and drying cycles. Hardly any petrified fibres could be observed in the aged autoclaved composite. Thus, the overall small change in the microstructural characteristics is in agreement with the observation that the changes in the mechanical properties during ageing were relatively small. It should be noted that the non-aged composite was rather dense to start with and not much densification or petrification took place on ageing. This behaviour may be associated with the greater stability of the autoclaved matrix and probably also with a different mode of carbonation in this matrix. In order to resolve these aspects, a detailed study of the mechanisms in the autoclaved matrix is required and in turn to be compared with carbonation of the normal cured cement matrix.

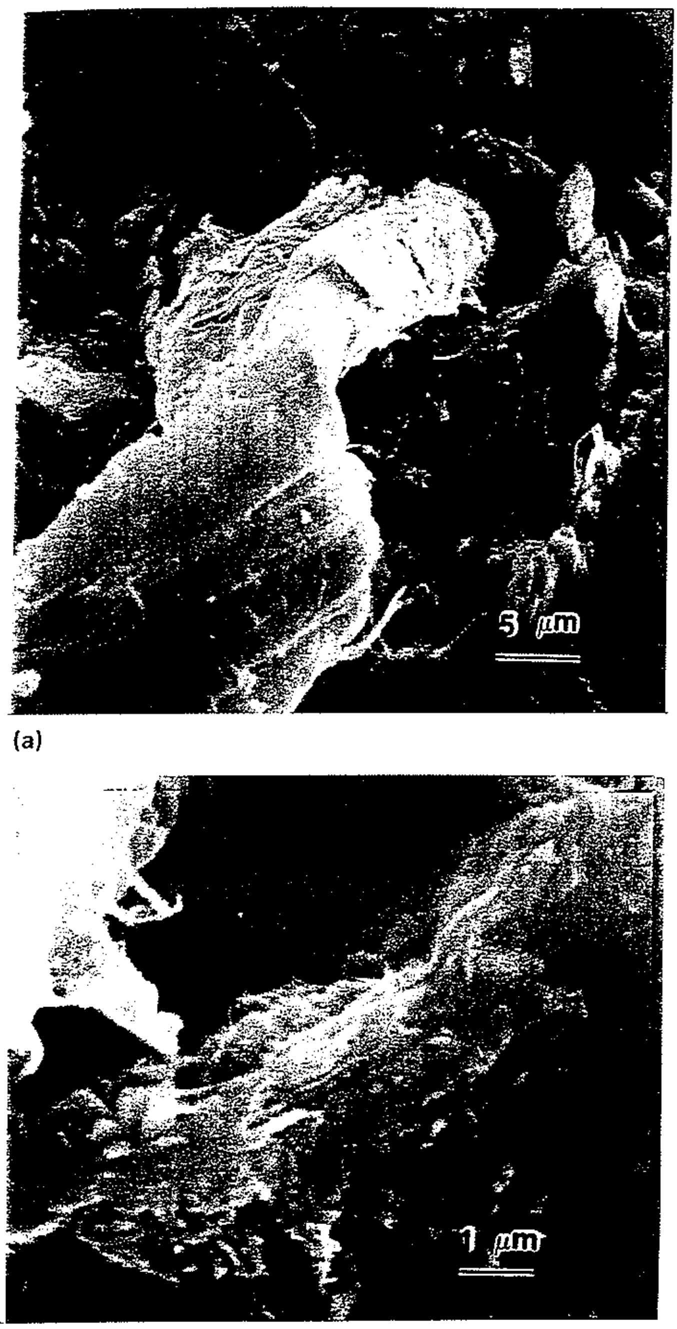


Figure 7 (a) Pull-out with helical fibre twisting in an accelerated aged autoclaved composite; (b) the dense matrix and gap to the right of the fibre in (a)

The trends observed here are in some respects similar to those reported by Sharman and Vautier [3, 4] but differences can also be noted. Sharman and Vautier [3, 4] also found that natural ageing and laboratory ageing in a CO_2 environment led to an increase in flexural strength and internal bonding. It is suggested that this change in mechanical properties is the result of carbonation, although they did not provide an explanation for this effect. Their conclusion was based on the study of an autoclaved composite.

CONCLUSIONS

1. The ageing of the autoclaved composite did not result in marked microstructural changes, and in particular little petrification of the cellulose fibres was observed, in spite of the fact that the composite had undergone carbonation.

2. This may account for the fact that the flexural strength and E-modulus of these products did not change much during weathering, and did not show the increase in flexural strength typical of a normal cured material.

3. The relative absence of petrification of fibres despite the high degree of carbonation may be the consequence of a different carbonation mode, which does not result in redistribution of the hydration products and their deposition in the cellulose fibre.

REFERENCES

1. Bentur, A. and Akers, S. A. S., 'The microstructure and ageing of cellulose fibre reinforced cement composites cured in a normal environment', The International Journal of Cement Composites and Lightweight Concrete, Vol. 11, No. 2, May 1989, pp. 99–109.
2. Akers, S. A. S. and Studinka, J. B., 'Ageing behaviour of cellulose fibre cement composites in natural weathering and accelerated tests', The International Journal of Cement Composites and Lightweight Concrete, Vol. 11, No. 2, May 1989, pp. 93–7.
3. Sharman, W. R. and Vautier, B. P., 'Accelerated durability testing of autoclaved wood fibre reinforced cement sheet composites', Durability of Building Materials, Vol. 3, No. 3, April 1986, pp. 255–75.
4. Sharman, W. R. and Vautier, B. P., 'Durability studies of wood fibre reinforced cement sheet', in Developments in Fibre Reinforced Cement and Concrete (eds R. N. Swamy, R. L. Wagstaffe and D. R. Oakley), Proceedings of the RILEM Symposium, Sheffield, 1986, paper 7.2.