

## **SELECTED ISSUES OF FIBRE-CEMENT SUSTAINABILITY: WOOD PULP REFINING ENERGY AND ACCELERATED AGEING OF FIBRES AND COMPOSITES**

DE LHONEUX, B., ALDERWEIRELDT, L., BOERSMA, A.\*, BORDIN, R., SAENEN, W., VAN DER HEYDEN, L., VERLEENE, D.

*Redco NV (R & D center of Etex group SA), Kuiermansstraat, 1, B-1880 Kapelle-op-den-Bos, Belgium;*

*\*TNO Science & Industry, De Rondom 1, P.O. Box 6235, NL-5600 HE Eindhoven,*

*The Netherlands*

---

### **ABSTRACT**

Sustainability has become an essential part in the value assessment of products and technologies. In this context, the present study investigates the reduction of energy used for wood pulp refining through an optimisation of operational parameters, particularly refining intensity. In view of the influence of this parameter on fibre length, a compromise has to be achieved between energetic efficiency and fibre quality. As product durability is an equally important aspect of sustainability, we further report data from accelerated and natural ageing of fibre-cement products with emphasis on the stability of synthetic fibres and on the carbonation of the cementitious matrix. It is shown that tenacity and molecular weight of PVA and PP fibres are stable in dry thermal ageing at 60°C. The rate of reduction of these two parameters at high temperature (110°C) is further discussed in terms of fibre service life at 25°C. These data are consistent with the observed physico-chemical stability of the fibres during natural exposure. Finally, we show that the rate of carbonation during natural weathering is initially fast and that it slows down with time. A similar trend is observed during accelerated ageing, particularly according to the "BAC" procedure (dry/wet cycling with CO<sub>2</sub> addition), confirming its validity. It is suggested to further develop accelerated ageing test methods for both fibres and composites in order to improve the predictability of fibre-cement property evolution during natural weathering.

### **KEYWORDS**

Cellulose, refining, PVA, PP, fibre, sustainability, carbonation, ageing, weathering

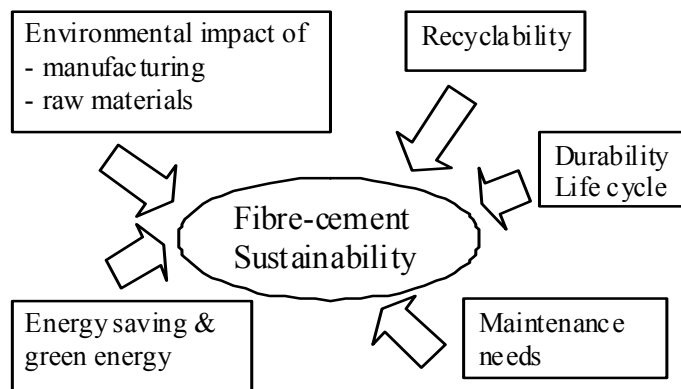
### **INTRODUCTION**

In the context of known global challenges such as the depleting of non renewable raw materials and energy sources or the growing environmental impact of human activities, increased attention is rightly given to the sustainability of industrial activities. Building products and fibre-cement, in particular, are no exceptions.

In this context, several issues can be addressed such as the environmental impact of raw materials production and of fibre-cement manufacturing, the durability and service life of the products, their needs for maintenance, their recyclability as well as their integration into building systems of high energetic efficiency and/or contributing to the production of green energy (Fig. 1).

We discuss in the present study two aspects of this sustainability issue: the reduction of the amount of energy used for the refining of wood pulp fibres and the evaluation of fibre-cement durability during accelerated ageing and natural weathering.

In the first part, we show that the minimum energy required to achieve a given freeness is dependent on refining intensity, expressed by the specific edge load, and on the fibre type. As fibre length decreases with refining intensity, a compromise has to be found between energy saving and fibre quality.



**Figure 1. Sustainability issues.**

In a second step, we investigate the stability of polymer fibres (polyvinylalcohol and polypropylene) in fibre-cement materials submitted to prolonged dry heat and to natural weathering. Fibres molecular weight and tenacity are found to be stable in prolonged oven ageing of fibre-cement samples at 60°C. From the rate of property reduction at 110°C of polypropylene fibres stabilised using hindered amine light stabilisers (“HALS”), a service life at 25°C in excess of 100 years could be expected for this fibre, based on recent models of polyolefin oxidative degradation. This explains earlier published results where, polyvinylalcohol and polypropylene fibres extracted from fibre-cement products exposed on roofs for up to 18 years showed no measurable signs of degradation.

Finally, we compare the rate of matrix carbonation of fibre-cement materials during accelerated ageing and natural weathering. In both cases, carbonation appears fast in the beginning and slows down with time. A lower degree of carbonation is observed in the high density fibre-cement (slates) than in the lower density one (corrugated sheets). Details of the “BAC” accelerated ageing test method could be further optimised in order to achieve in a shorter time the degree of carbonation observed on roofs after several years on roofs.

## ENERGY CONSUMPTION IN WOOD PULP REFINING

Wood pulp fibres are usually refined in the process of fibre-cement manufacturing. This operation is needed for process reasons and sometimes for product performances as well. It renders the fibres more flexible and conformable due to fibre wall delamination and fibrillates their surface. Simultaneously, fibres are shortened and fines are created. Degree of refining is usually expressed by the Schopper Riegler scale (°SR) or by the Canadian Standard Freeness (mL).

The refining process is energy intensive and specific energy consumption of up to 15 kWh/MT/°SR are not unusual. The total energy needed obviously depends on the freeness, but in case of low freeness, up to 500 - 800 kWh/MT can be needed. This represents not only a significant cost, but also a heavy environmental impact.

Refining trials were organised using a 20” double disc refiner and two wood pulps, i.e., an unbleached and a bleached softwood kraft pulp both of radiata pine. The refiner was operated in a tank recirculation mode, i.e., pulp was recirculated to the feeding tank until the target °SR was achieved. Three refining intensities were investigated: 1, 2 and 3 Ws/m, using the following plate design: 4/4/7,1 (all in mm) and +10° master angle. Calcium hydroxide saturated water was used for repulping the fibres in order to reflect the alkalinity of process water in fibre-cement mills.

The intensity dependence of the specific energy needed for refining the pulps up to 60°SR is shown in Fig. 2. For the unbleached pulp, a minimum is observed around 2 Ws/m. For the bleached pulp, the energy decreases almost linearly with increasing intensity.

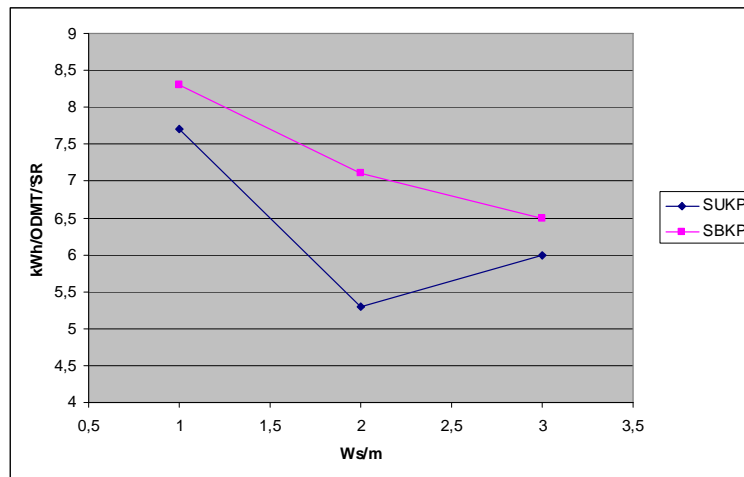


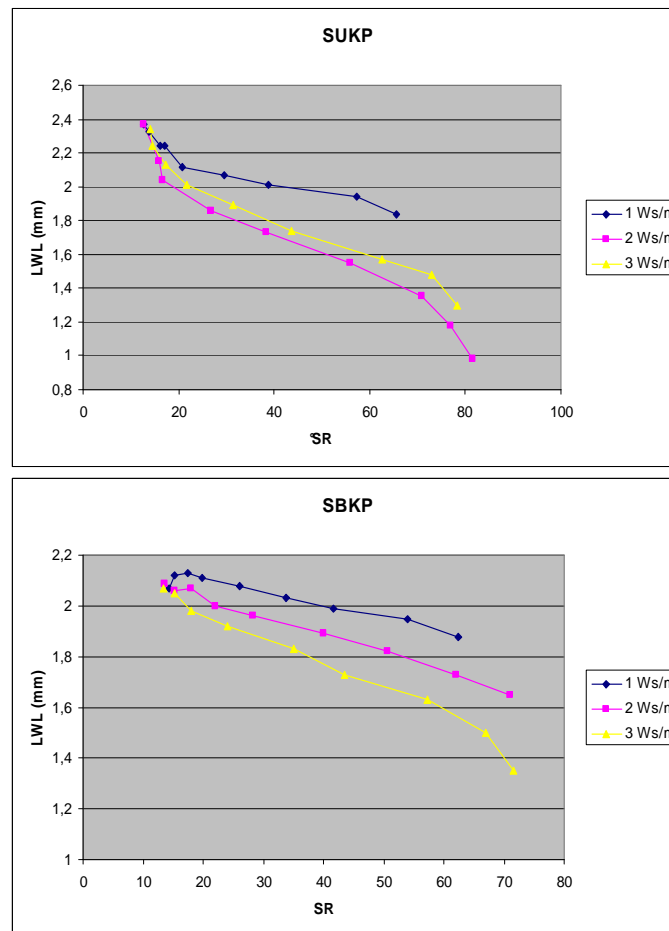
Figure 2. Freeness related specific energy versus refining intensity.

Fibre length decreases with increasing °SR (Fig. 3), as expected, and with increasing refining intensity, except for the unbleached pulp beyond 2 Ws/m. For this pulp, high intensity refining results in less fibre shortening than at medium intensity.

The respective evolution of length and energy is consistent with the assumption that cutting the fibres is energetically less expensive than fibrillating them. Further, the observation of a minimum in the energy versus intensity curve is also consistent with recent models of the refining process (Joris 2007, Bordin 2008). There remains however uncertainty as to how to calculate the refining intensity. In the present study, we express the refining intensity or specific edge load according to the classical edge length theory, but the same recent models recommend to include bars angle into the intensity calculation (“MEL”, Meltzer 1995 ) or to express it in terms of number of crossing points (Roux et al. 2007). These parameters as well as the freeness dependence of the energy minimum shall be integrated in future studies.

On the other hand, there is no clear explanation for the fact that the energetic optimum occurs at a lower intensity for the unbleached than for the bleached pulp. But this indicates that refining operational conditions need to be adapted to each pulp.

Finally, as refining intensity strongly influences fibre length, a compromise has to be achieved for each pulp furnish between energy saving and fibre quality.



**Figure 3. Relation between fibre length (LWL) and degree of refining at three refining intensities for an unbleached (SUKP) and a bleached (SBKP) softwood kraft pulp.**

## THERMAL AGEING OF SYNTHETIC FIBRES

Long term performance of building products in their intended application is an important aspect of their sustainability. Fibre-cement materials are used in a wide range of applications such as roofing, cladding, partitioning, ceiling, flooring, etc... Environmental conditions are particularly severe on roofs: temperature and humidity widely fluctuate during the day and across the seasons, numerous frost-defrost cycles are encountered in northern latitudes or at medium and high altitude, intensive UV irradiation is present in many places. For example, temperatures recorded on roofs in the Belgian highlands (the Ardens, 400 – 600 m altitude) fluctuate from below  $-20^{\circ}\text{C}$  up to  $70^{\circ}\text{C}$ . Correct product design has to prevent premature product failure by, e.g. fibres degradation, excessive bonding of fibres to cement, interlayer delamination, macrocracking, etc...

Several studies have shown the stability of polyvinylalcohol (PVA) and polypropylene fibres (PP) in the course of accelerated & natural weathering (Akers *et al.* 1989, Hannant 1998, Lhoneux *et al.* 2002 & 2004, Nishiyama *et al.* 2006). PVA fibres have been successfully used in roofing applications for more than 25 years. Comparatively little information has been published on the stability of these fibres under prolonged exposure to dry heat, as can occur on roofs in summer. Therefore, laboratory made fibre-cement samples reinforced by PVA and by PP fibres were aged in ventilated ovens at  $60^{\circ}\text{C}$  and  $110^{\circ}\text{C}$ . The PVA fibres were provided by Kuraray, Ltd and the PP fibres were supplied by Daiwabo, Ltd, both from Japan. The PP fibres contained 0.3% of hindered amine light stabiliser ("HALS", Chimisorb 944 from Ciba Specialty Chemicals). The fibres were extracted from the samples after softening/dissolution of the cement matrix using diluted

acid. Tenacity at break of the fibres was measured using the Fafegraph single fibre tester from Textechno GmbH, assuming the nominal fibre titer to convert the force measurement into tenacity. In the case of the PVA fibres, the measurements were made by Kuraray, Ltd.

As can be seen from Fig. 4, prolonged thermal ageing at 60°C of the fibre-cement samples has hardly any influence on the fibre properties in the time frame investigated.

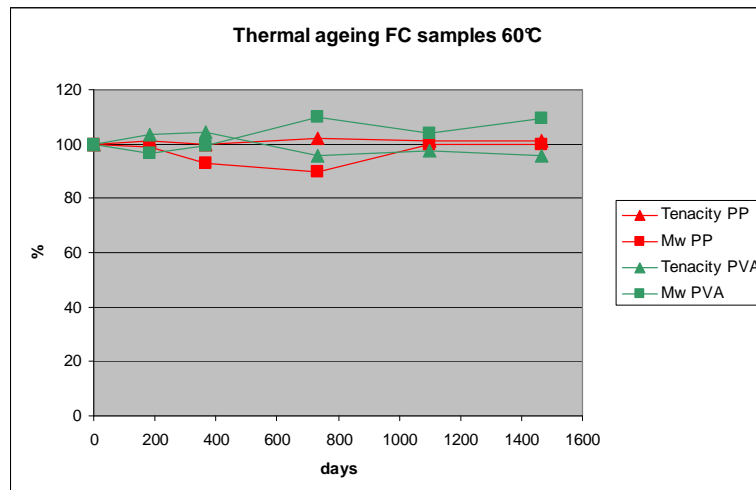


Figure 4. Tenacity and molecular weight of PVA & PP fibre samples extracted from fibre-cement samples thermally aged at 60°C in a ventilated oven.

At higher temperature (110°C), tenacity and molecular weight reduce for both fibres as can be seen from Fig. 5. In the case of PVA fibres, tenacity is no longer measurable after 1 year and in the case of PP, molecular weight reduces with time in a curved plot, i.e., fast in the beginning and slowing down afterward, following apparently a first order kinetics. For this fibre, tenacity reduces at a slower pace in the beginning but drops when Mw reduces below 60.000, i.e., at 40% of its initial value.

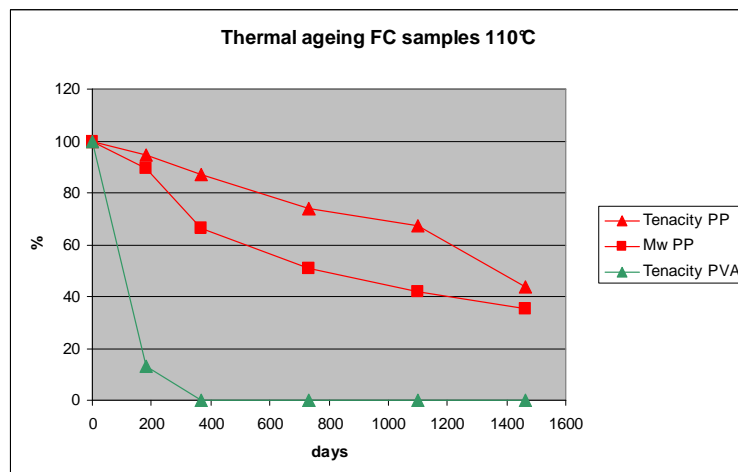


Figure 5. Tenacity and molecular weight of PP and PVA fibres extracted from fibre-cement samples exposed to thermal ageing at 110°C in a ventilated oven.

Polymeric materials undergo degradation reactions in the presence of oxygen. The oxidative degradation reactions are influenced by many factors such as temperature, stress, oxygen solubility and diffusion rate, concentration and diffusion of stabilizers, crystallinity, molecular orientation, polymer physical transitions, etc... From the 60°C data, it appears that the fibres embedded in the cement matrix are intrinsically stable at temperatures which can occur during natural weathering. Previous experiments showed that unfixed PP

fibres aged at 60°C in a ventilated oven did show some degradation, perhaps due to a faster stabiliser loss and/or to a fibre relaxation.

At the higher temperature of 110°C, both fibres degrade according to polymer specific features. For PVA, the degradation is rather rapid, as expected from literature data (Sakurada 1985). It can be explained by faster auto-oxidation reactions, due to the high chain mobility and permeability of oxygen at temperatures much higher than its glass transition temperature (70 – 80°). This degradation involves the formation of polyene structures, chain scission, cross linking, etc.... The extrapolation of the degradation to lower temperatures is not possible due to this phase transition. For the polypropylene fibres, the auto-oxidation reaction appears rather slow in comparison with some results from thermal ageing of geotextiles (Mueller et al. 2003). On the other hand, high stability was also observed by Hannant (1998) in thermal ageing of nets. In the first case, an efficient stabiliser package and a high level of molecular orientation were considered as key elements of long term stability. The fibres used in the present study have a tenacity twice as high as those investigated by Mueller et al. (2005) and their molecular orientation is also higher. Additionally, the fibres of the present study were stabilised using the hindered amine light stabilisers (“HALS”) which are known to be excellent long thermal stabilisers.

The extrapolation to lower temperature, e.g. 25°C, of the degradation speed at 110°C needs some assumptions regarding the temperature dependence of the reaction rate, the oxygen diffusion rate, polymer relaxation, diffusion and degradation rate of the stabilisers, etc... As far as the temperature dependence is concerned (Woo et al. 2001), recent data show the logarithm of the PP service life versus the reciprocal of the temperature to be a curved plot instead of a straight line as could be expected from an Arrhenius relation. Using the model curve mentioned in the a.m. reference ( $\text{Log } t = -35,536 + 2,666 e^4(1/T) - 3.993 e^6(1/T)^2$ ), and discounting all other factors, the service life at 25°C of the fibre, defined as the time to 50% property reduction would amount to about 150 years for molecular weight. This is of course an oversimplification and many additional factors need to be taken into account for assessing the service life on site, such as the influence of water on the diffusion of oxygen, the loss of stabiliser to the environment, the eventual presence of stress in the fibre, etc...Such aspects should be further investigated in order to provide an accurate prediction of fibre and product service life in site. In any case, the choice of a suitable stabiliser such as HALS is important for the thermal stability as is shown from the following data from oven ageing of filaments:

Fibre	Time to brittle failure (110°C)
Unstabilised	19 days
0.3% chimasorb 944	349 days

Finally, these results also explain why no significant tenacity and molecular weight change were observed in fibres extracted from naturally weathered fibre cement samples (Lhoneux et al. 2004).

## MATRIX CARBONATION: ACCELERATED VS NATURAL AGEING

Besides fibre stability, long term performance of fibre-cement has also to take into account the evolution of the cementitious matrix. Atmospheric CO<sub>2</sub> reacts with portlandite (calcium hydroxide) and with hydrated cement phases (amorphous calcium silicate hydrates) forming calcium carbonate. It is a complex reaction involving several steps, such as the dissolution of CO<sub>2</sub> in water, the dissociation of H<sub>2</sub>CO<sub>3</sub> and of Ca(OH)<sub>2</sub>, the reaction of calcium and carbonate ions and the formation of water (Thiery et al. 2005). The in-situ formation of calcium carbonate increases product density, stiffness, bending or tensile strength and lowers dimensional instability and strain at break (Kalbskopf et al. 2002). It reduces porosity and slows down liquid and vapour transports. It also reduces the alkalinity of pore water.

In order to assess the rate of carbonation during natural weathering and accelerated ageing, we measured in a first step the density evolution of medium density ( $1.67 \text{ g/cm}^3$ ) fibre-cement samples submitted to the "BAC" test ("Beschleunigte Alterung mit  $\text{CO}_2$ ") which consists in wet/dry cycling during which  $\text{CO}_2$  is added in the drying phase. The cycling time was shortened from the original sequence published by Akers and Studinka (1989) and consisted of the following steps:

- (1) submerged under water at ambient temperature during 5 hr;
- (2) drying at  $65^\circ\text{C}$  during 0.5 hr;
- (3) cooling during 0.5 hr;
- (3): air(78%)/ $\text{CO}_2$  (22%) circulation at ambient temperature during 3 hr;
- (4): drying at  $65^\circ\text{C}$  for 3 hr.

Simultaneously, we similarly followed-up the density evolution of two series of corrugated sheets (unpressed, density before ageing =  $1.53 \text{ g/cm}^3$  and pressed, density before ageing =  $1.6 \text{ g/cm}^3$ ) exposed to natural weathering in Elsenborn, Belgium (about 500 m altitude). As appears from Fig. 6a, the density increases fast during the early phases of the BAC test and slows down afterwards. After 1500 to 2000 cycles, the density has increased by 0.12 to  $0.14 \text{ g/cm}^3$ . During natural weathering, density similarly increases initially fast, and slows down after a few years. For these sheets, the density increase from 5-10 years natural weathering ( $0.14 \text{ g/cm}^3$ ) appears similar to that of samples exposed to 1500 to 2000 BAC cycles. More scatter is observed in the data from the natural ageing series, presumably due to inhomogeneities within sheets, or between sheets from the same lot. The BAC test sequence could be further optimised in order to achieve in a shorter time the rate of carbonation observed in, say, 10 years old sheets. In particular, the moisture content of the sheets at the beginning of the carbonation sequence is critical for reactions to occur rapidly. Also, the water produced by the carbonation has to be removed in order to allow the reactions to proceed further. Dimensions and density of the samples need also to be taken into account, as they influence the time needed to achieve equilibrium moisture content in the various phases of the test.

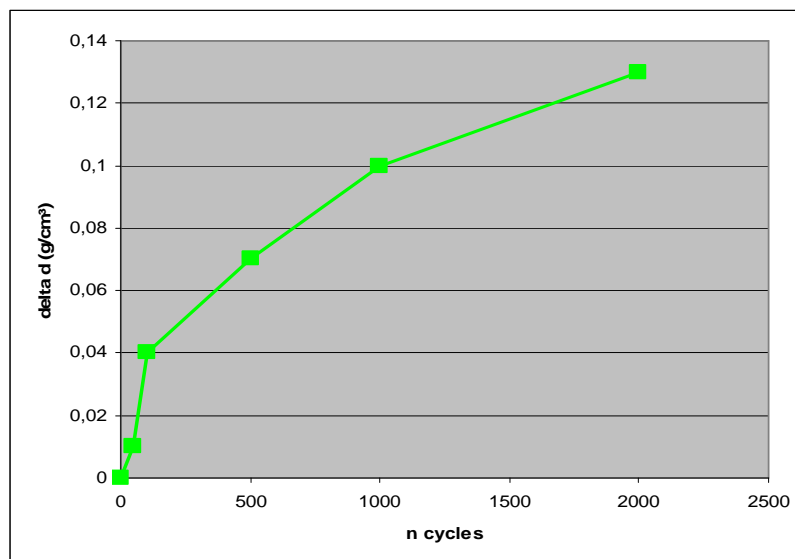


Figure 6a. Density increase of fibre-cement samples exposed to prolonged BAC cycling.

In an additional series of experiments, we compared the carbonation rate of corrugated sheets and of slates exposed on the one hand to a 100%  $\text{CO}_2$  atmosphere for up to 72 hr, and on the other hand to the same BAC test as described above. Simultaneously, in situ formed carbonate content was also determined on products retrieved from natural exposure sites in Belgium (Elsenborn) and France (Vitry-en-Charolais). As can be seen on Fig. 7, the degree of carbonation achieved in corrugated sheets after 116 and 226 BAC cycles exceeds that achieved by a 72 hr exposure to  $\text{CO}_2$  but remains inferior to that of 6-10 years old sheets.



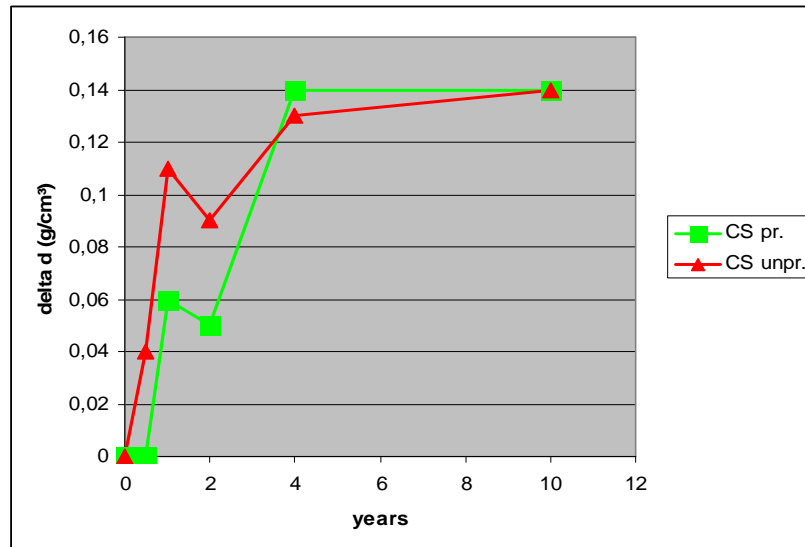


Figure 6b. Density increase of fibre-cement corrugated sheets exposed to natural weathering in Elsenborn, Belgium (CS pr. = pressed corrugated sheet, CS unpr. = unpressed corrugated sheet).

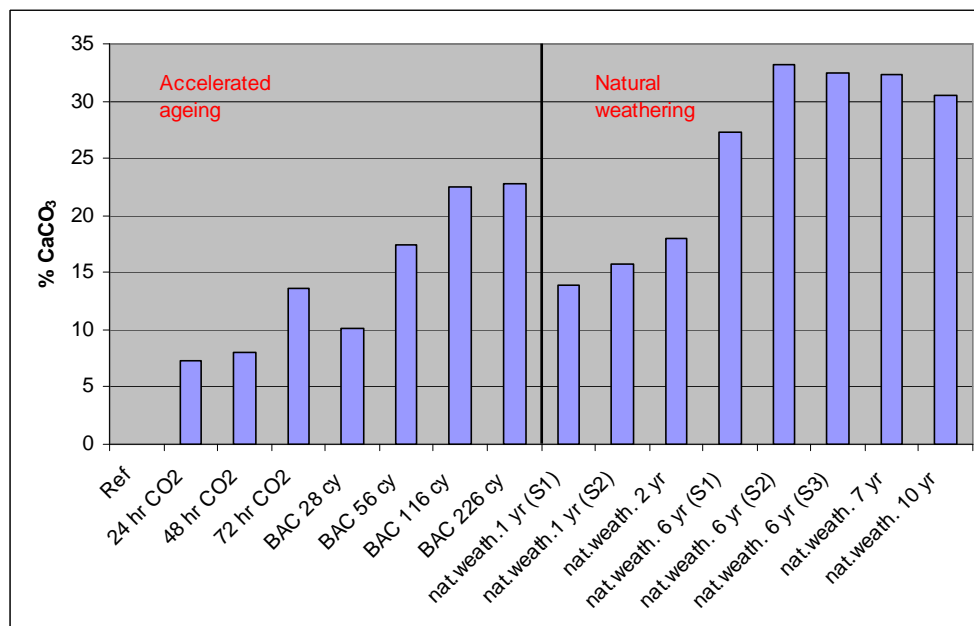
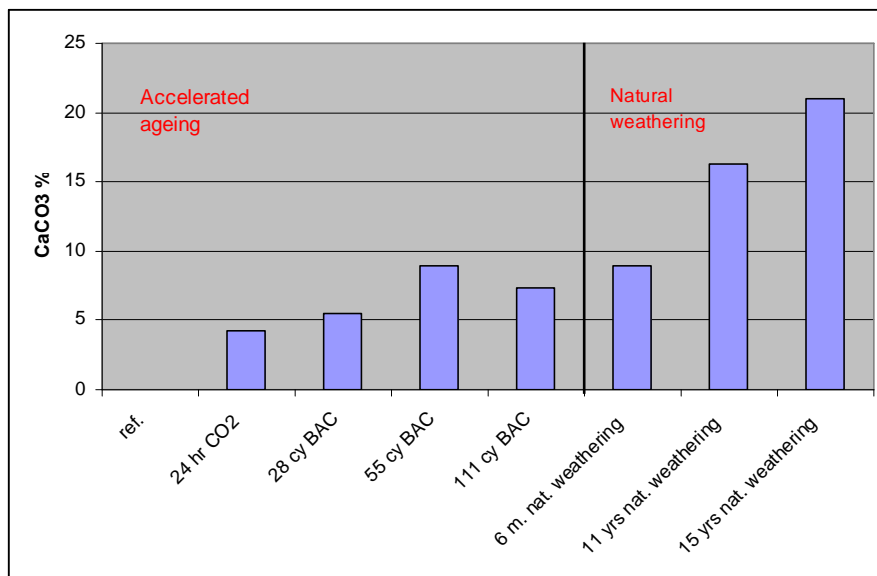


Figure 7. Calcium carbonate content of corrugated sheets after accelerated carbonation tests and natural weathering in Belgium.

Within the same product, the degree of carbonation can be heterogeneous with areas fully carbonated, while other areas have a colour reaction to the phenolphthaleine test. This has been observed in some corrugated sheets characterized by a carbonation “front”, or in others where differently carbonated areas coexist, such as tops and valleys of the corrugations. Such differences may result, at least partially, from differences in moisture content which is a key element for the carbonation reactions.

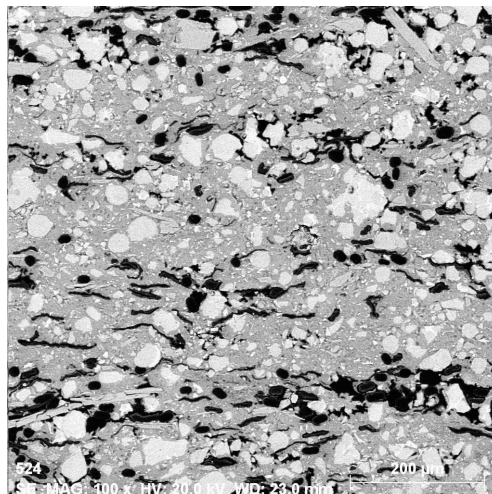
For slates, although the sampling is more limited, it seems that the amount of CO<sub>2</sub> uptake is less important than for corrugated sheets (Fig. 8).





**Figure 8. Calcium carbonate content of slates after accelerated carbonation tests and natural weathering in France.**

The initially rapid and subsequent slower carbonation rate appears consistent with the model where portlandite crystals are progressively surrounded by rims of calcium carbonate microcrystals forming a compact coating through which further ion transfer progresses with more difficulty (Thierry et al. 2005). A direct carbonation of the CSH phase is also known although the microstructural details are less well documented. The uptake of CO<sub>2</sub> decreases product porosity, slowing down water and vapour transport and thus carbonation reactions. Even after 15 years of natural weathering in the French exposure site, only a fraction of the clinker of the slates has reacted as can be seen from Fig. 9, indicating that hydration and carbonation can still continue although the rate of these reactions is likely to be low. The low cement hydration and carbonation levels in these products may result on the one hand from the low initial water/cement ratio resulting in a reduced hydration level, and from the low water permeability and diffusion rates in high density matrices.



**Fig. 9. SEM image in backscattered mode of a slate exposed to 15 years of natural ageing. White areas represent unhydrated clinker particles.**

Similarly, the presence of a coating on one or both sides of the products reduces the rate of water diffusion and thus the rate of carbonation.

## CONCLUSIONS

Fibre-cement sustainability requires an efficient use of energy for manufacturing, including for cellulose fibres refining. Pulp specific operational parameters have to be optimised, particularly refining intensity. A compromise between energetic efficiency and fibre quality has to be achieved in view of the respective energetic needs of fibrillation and cutting. Sustainability also implies satisfactory long term performance of products on roofs, what requires the availability of accelerated ageing methods in order to assess physico-chemical changes in fibres and in cementitious matrices. Data from the thermal ageing of fibres confirm their long term stability at 60°C while we discuss rate of property reduction at 110°C in terms of service life at 25°C. These data are consistent with the observed physico-chemical stability of synthetic fibres during natural weathering. Further, a similar carbonation rate evolution is observed in samples submitted to the "BAC" accelerated ageing and during natural weathering, i.e., fast in the beginning and slowing down afterward. Accelerated ageing test methods for both fibres and matrix should be further optimised in order to provide fast and accurate predictions of fibre-cement property evolution during natural weathering.

## LITERATURE CITED

- Akers, S., Studinka, J. 1989. "Ageing behaviour of cellulose fibre cement composites in natural weathering and accelerated tests." *International Journal of cement Composites and Lightweight Concrete*, 11, 2, 79-91.
- Akers, S., Studinka, J., Meier, P., Dobb, M., Johnson, D., Hikasa, J. 1989. "Long term durability of PVA reinforcing fibres in a cement matrix." *International Journal of cement Composites and Lightweight Concrete*, 11, 2, 79-91.
- Joris, G. 2007. "Gestion optimale d'une unité industrielle de raffinage." *Revue ATIP*, 61(2), 6-18
- Bordin, R. 2008. "Modélisation physique du fonctionnement d'un raffineur de pâte à papier." Institut National Polytechnique de Grenoble thèse de doctorat
- Hannant, D.J. 1998. "Durability of polypropylene fibers in Portland cement-based composites: 18 years of data." *Cement and Concrete Research* 28,12,1809-17.
- Kalbskopf, R., Lhoneux (de), B., Van der Heyden, L., Alderweireldt, L. 2002. "Durability of fiber-cement roofing products." 8th IIBC conference, Sun Valley, Idaho.
- Lhoneux, B. (de), Akers, S., Alderweireldt, L., Amyia, S., Carmeliet, J., Hikasa, J., Saenen, W., Studinka, J., Tomka, I., Vanden Bosch, M. 2002. "Durability study of PVA fibres in Fibre-cement products." *Proceedings of the Ivth International symposium on concrete for a Sustainable Agriculture*, Ghent (Belgium).
- Lhoneux, B. (de), Alderweireldt, L., Albertini, E., Capot, Ph., Honorio, H., Kalbskopf, R., Lopez, H., 2004. "Durability of polymer fibres in air-cured fibre-cement roofing products." 9<sup>th</sup> IIBC conference, Vancouver, Canada
- Meltzer, F. P. 1995. "Diminution du besoin en travail spécifique de raffinage, grâce à un nouveau chiffre caractéristique et à des garnitures en matériaux d'un genre nouveau." 48<sup>ème</sup> congrès ATIP, Bordeaux, France.
- Nishiyama, M., Yamamoto, R., Hoshiro, H. 2006. "Long term durability of Kuralon (PVA fibre) in alkaline condition." 10th IIBC conference, Sao Paulo.
- Roux, J.C., Bloch, J.-F., Nortier, P., 2007. "A kinetic model for pulp refining, including the angular parameters of the equipments." *Appita J.*, 60,1,29-34
- Sakurada, I. (1985) *Polyvinyl Alcohol Fibres*, Marcel Dekker, Inc, New York
- Thierry, M., Dangla, P., Villain, G., Platret, G. 2005. "A prediction model for concrete carbonation based on coupled CO<sub>2</sub>-H<sub>2</sub>O-ions transfers and chemical reactions. 2005." 10DBMC international conference on durability of building Materials and components, Lyon, France.
- Woo, L., Khare, A.R., Sandford, C.L., Ling, M.T.K., Ding, S.Y., J. 2001. "Relevance of high temperature oxidative stability testing to long term polymer durability." *Thermal Anal. Cal.* (2001) 64, 539 – 548.
- Zweifel, H. 1998. "Stabilization of Polymeric Materials." Springer-Verlag Berlin Heidelberg New York

## ACKNOWLEDGEMENTS

The work of Kuraray Ltd for tests on PVA fibres from aged products is gratefully acknowledged.

---