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Session 7: Dr H. G. Higgins

THE reaction between paper and its environment is by definition the ecology of paper. This word was once understood only by biological scientists and scholars of Greek, but it is now on everybody's lips, so session 7 should have proved immensely popular. Of course, we have largely confined our considerations to the response of paper to its environment or to imposed conditions of temperature, humidity and mechanical or chemical stress; we have not been too concerned today with the way in which the total environment, as it involves man, is affected by paper and by the technological processes that constitute papermaking—or more broadly, the pulp and paper industry. It is as well to note, however, the connection between paper's response to and its effect on the environment, if only to ensure, as the sledgehammers of change swing ever faster, that we do not lose sight of the relevance of our discussions to the current pressing problems on the interface between the paper industry and the rest of society. Nor, we hope, will the relevance be lost on those responsible for sponsoring our attendance at the next symposium.

As this summing-up comes so soon after the session itself, I shall have to be honest with you and admit that certain of the matters I shall mention have been considered in advance, so that this is in the nature of a preview delivered after the event.

The question of permanence was raised by Dr Hudson and in his very useful written dissertation—which I imagine is just what publishers, librarians and archivists and the technologists who advise them need to know about this subject—he clarified for us the distinction between permanence and durability. The other day, I had the privilege of visiting the ancient city of Ephesus and, sitting and admiring the beautiful lines of Hadrian's temple, I took the opportunity (as one tends to do on such occasions) to reflect on the meaning of permanence. Marble is not a particularly durable material, except in protected situations, yet it admirably fulfilled its role in this and other cultures. One wonders if the rulers of the time were more concerned with permanence or temporal glory and what mechanical and chemical tests were carried out by the Ephesian architects and builders to determine how their structures would withstand the onslaught of the centuries. Perhaps the whole concept of permanence has relevance only within the framework of a particular culture. In the paper field, does not the seeking of extreme permanence

presuppose a continuation of present technologies? Will future generations store all their archives in supercomputers?—or, for that matter, will they have any interest in history?—even in reading?

Nevertheless, we must provide for a continuation of our own culture in terms of a projection of our technological trends. In this respect, the papers presented this morning have been extremely valuable. From Hudson's paper, the factors favouring permanence emerge clearly as relatively high pH value, low temperature and the nature of the fibrous furnish. Methods are now available for sizing under neutral or alkaline conditions and it is forecast that this will soon be possible without an undue increase in cost. The use of usably alkaline fillers such as calcium or barium carbonate can also improve permanence. Because the importance of acidity has been questioned, Hudson has done well to re-analyse the results of Dixson & Nelson and show that there is indeed a good correlation between pH value and ageing. It was interesting to hear that whole books can now be treated to make them more permanent—for example, with magnesium methoxide dissolved in dichlorodifluoromethane. The effect of temperature is illustrated by the fascinating example of the book left in the snow of Antarctica in 1912. When even the protein of mammoths can be preserved in the ice for many millenia, it is not surprising that cellulose stands up well to the cold.

Another interesting point made by Hudson is that Mr Murray was complaining about the impermanence of paper in 1829, 25 years before chemical pulping of wood was introduced. I don't know whether it is possible, however, that some spruce groundwood, was beginning to find its way into paper furnishes about that time. The pulp order of rag, high alpha-cellulose, low alpha-cellulose, groundwood seems to be well established as that of decreasing paper permanence. The absence of unsaturated linkages in the β-1-4 glucan would seem to be a major reason for the stability of cellulose, but it is probable that the higher crystallinity of cotton cellulose (90 per cent) in comparison with purified wood cellulose (50 per cent) would lead to lower reactivity.

It might be thought that the presence of lignin (which contains conjugated systems) and of non-cellulose wood polysaccharides (which particularly in hardwood pulps may contain acetyl and carboxyl groups) would accelerate the ageing process. The removal of lignin by oxidative bleaching has been reported to lead to superior ageing properties. Luner & Cardwell, however, have shown in their experiments on the thermal stability of papermaking pulps under conditions of accelerated ageing that the presence of lignin or high hemicellulose content was not of itself sufficient to cause rapid ageing. Luner had previously shown that the degree of bonding in paper influences the chemical reactions occurring at the contact areas, which lead to cross-linking between fibres and to ageing effects. (This appears to be crudely

analogous to the ageing process in organisms such as *Homo sapiens*, where protein cross-linking is a major effect, if not the essence of the process. It has even been suggested that the discovery of specific enzyme systems to inhibit cross-linking may be the key to eternal youth.)

Luner & Cardwell see the loss of mechanical properties on accelerated ageing as the result of two competitive structural effects, one a slow loss of fibre strength and the other a marked increase in interfibre and intrafibre bonding. Folding endurance appears to be the best index of ageing under accelerated conditions and a useful procedure has been developed whereby different loads may be used appropriate to the state of the paper and the fold results converted to the same basis by the use of a linear correlation between the logarithms of the load used in the test and of the folding endurance. Conditions for accelerated ageing tests must be carefully chosen for proper simulation and it is indeed surprising that the standard U.S. tests make no provision for moisture in the sheet. If the mechanism of degradation is to be the same, the temperature chosen should presumably not be above the glass transition point of any of the sheet constituents. The use of the Arrhenius equation implies no change in mechanism with temperature and a constant activation energy.

A number of practically useful points were made by Luner & Cardwell: sulphite pulps can be more permanent than kraft pulps, although weaker initially; pine kraft pulps are more stable than those from birch. A weak semi-chemical birch sulphite pulp showed very good ageing properties, possibly because of higher pH value, whereas an initially strong, bleached birch kraft pulp aged very poorly. The conclusion is reached that the most useful permanent papers will be produced from pulps with high fibre strength and without excessive initial fibre bonding.

The important paper by Silvy & Le Nest dealt with the development of chromophores and, by using diffuse reflection spectroscopy, they were able to determine the absorption bonds that appear when cellulose is heated in the dark. The colour development was shown to be reversible when the cellulose was exposed to daylight at ambient or low temperatures. These photo-restoration effects are very interesting and can be used for obtaining a higher degree of whiteness than in the initial state. It is interesting to note that the observed phenomenon takes place both for lignified cellulose such as newsprint and for pure cellulose. Silvy's work is relevant to the choice of conditions for the study of accelerated ageing under standard conditions, as well as to the response of paper products to the natural environment, from the points of view of both increasing permanence and of sponsoring degradation.

Turning from permanence to durability, Dr Graminski presented us with an interesting and straightforward account of his experiments on the flexing

of paper. It seems likely that, in these days of data cards and documents that require automatic handling, his new apparatus will find ready application and be added to the armoury of paper tests. This is all the more likely in that he could find no correlation between durability as measured with his apparatus and any other paper property measured. It is interesting to note that tearing strength, which we know to be associated primarily with fibre length and strength, is least affected by flexing, whereas bonding properties are far more susceptible. That the interfibre matrix does in fact deteriorate on flexing was shown directly by the scanning electron microscope, cracks developing in a film-like matrix consisting presumably of microfibrils produced by beating. The beautiful stereoscopic pictures shown by Dr Graminski really allowed us to look deeply into his materials. It would be of considerable value to have data of flexing for papers made from chemical pulps produced by various processes and with a range of morphological characteristics. Newsprint appears to be far less durable than rag papers, as expected, but a comparison between high quality chemical pulp and rag pulps would be most instructive, softwood and hardwood pulps could also be compared. In addition to the applications mentioned, Graminski's study is relevant also to the question of the suitability of different grades of paper for banknotes.

The final paper in this session was presented by de Ruvo, Lundberg, Martin-Löf & Söremark on the influence of temperature and humidity on the elastic and expansional properties of paper and the constituent fibres. This contains a wealth of interesting scientific information that should be of particular relevance to those concerned with the use of paper and board for packaging purposes, for example, in corrugated fibreboard containers. In this connection, I wonder to what extent other countries experience the problem we have in Australia of getting the converting industries to apply the results of basic research as readily as does the papermaking sector. De Ruvo *et al.* show that the single fibres and papers made from them respond to humidity and temperature in a similar fashion. An important conclusion, perhaps common to anisotropic systems, be they magnetic fields, crystals or fibre assemblages and brought out by Steenberg & Rance in their early studies on the rheology of paper, is that you cannot get anything for nothing, so that improvements in a property in one direction are accompanied by a deterioration in another. The isotropic sheet may be considered to be the system of maximum entropy, whose properties in respect to both dimensional instability and elasticity are the geometric means of those in the machine and cross directions. Other unifying generalisations are provided by De Ruvo *et al.* in that the product of the coefficient of linear thermal expansion and the elastic properties is constant in all directions and hygroexpansion follows a similar relationship.

In the paper physics Derby, it has always been very difficult to keep the Swedish Forest Products Research Institute in sight. Since I saw the work on fibre torsion at Syracuse last year, I have been resolved to attempt applying this technique to some of our problems; de Ruvo *et al.* have already made use of the method to show that fibre swelling is proportional to moisture regain and have suggested that the measure of twist could be used to determine fibrillar angle. The enhanced creep rate observed under conditions of changing moisture content was, I believe, first described for wood by Armstrong & Kingston; since then, considerable effort has been expended on its theoretical explanation. It is a most important phenomenon for the practical utilisation of papers and board, for example, in relation to the behaviour of stacked fibreboard fruit containers being transported from Australia to England.

If I may end on another somewhat controversial point; the higher sensitivity of bleached fibres to temperature and humidity changes compared with unbleached fibres should lend support to those who view with some disquiet the tendency to produce packaging materials that have undergone more processing than is warranted by the function they are called on to perform.

I know I have not given a proper appraisal of the valuable remarks from the floor such as the useful contributions of Back and of Balodis, but it has been difficult to do this at short notice.

The papers this morning fell into a coherent pattern and, for this, the Fundamental Research Committee is to be congratulated. I believe that, when they are published, the papers will continue for a long time to provide inspiration for future work, as the transactions of these previous symposia have done.