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THE FUNDAMENTAL PROBLEM IN RECYCLING

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Synopsis From a review of the literature and our own work, it appears that the problem requiring most urgent attention is the nature of secondary fibres themselves. It is shown that the essential difference between secondary and virgin fibres is their bondability and that this depends on the papermaking process by which the secondary fibres were generated. Mention is made of some treatments which go some way towards restoring the strength of sheets containing recycled fibre.

Introduction

PAPER is different from wood pulp as a raw material for the making of paper. The truth, or otherwise, of this statement raises the really fundamental question to be asked about the recycling of paper. There are, of course, many other aspects of the more immediate, practical issues in the recycling field. The economics and mechanics of collection, sorting, grading and storage is one such. The proper machinery and energy requirements for stock preparation from waste paper is another. The possible technical compromises between price, quality and production difficulties for grades incorporating waste paper is a third. Answers to these questions are appearing in a steady stream and now make an interesting, useful and large component of the literature of our technology. But, to return to our first statement; is it purely academic or is it important? In a paper at the Chester conference, Minshall[1] suggested that the practice and procedure for the re-use of waste paper is reaching a state where some new thinking is required. This, he suggests, comes about because the sources of high grade waste are nearly fully used in this country at present. The new sources, from which large quantities of waste paper could become available, are essentially sources of low grade waste; households and the like. At the same time, those grades of paper and board which, traditionally, are made from large proportions of waste, are approaching the 100 per cent waste furnish: ecological bliss! This means that, to use more waste, we need to find ways of incorporating it in the higher grades of paper, whereas

the additional waste that could become available is of the lowest grades. The solution to this problem will be partly economic; the better grades of waste may become priced out of the lower grade furnishes. We still need to go back to fundamentals in considering whether waste paper can ever become an adequate raw material for the making of higher grades of paper. If paper really is an inferior raw material for papermaking, and if its inferiority is so great as to make it impossible, very difficult or even more expensive to use in the making of higher grade papers, then we shall have great difficulty in increasing our overall re-use factor above, say, 0·5.

It is the purpose of this paper to examine the evidence for this statement and present it in a form to stimulate discussion. It may be considered under the following headings: 'The Results of Recycling experiments', 'The Effects of papermaking on Fibres' and 'The Effects of variation in Papermaking Procedure'.

The results of recycling experiments

THERE appears to be little doubt or disagreement among workers in this field that the classic recycling experiment always has the same, by now predictable, result. Fig. 1 is a typical example of results published by McKee,[2] Bovin, Hartler and Teder,[3] Cildir and Howarth[4] and Szwarcztajn and Przybysz.[5] There may well be others to whom I apologise for their omission from the list. From these results we see that, if we take some pulp, make of it

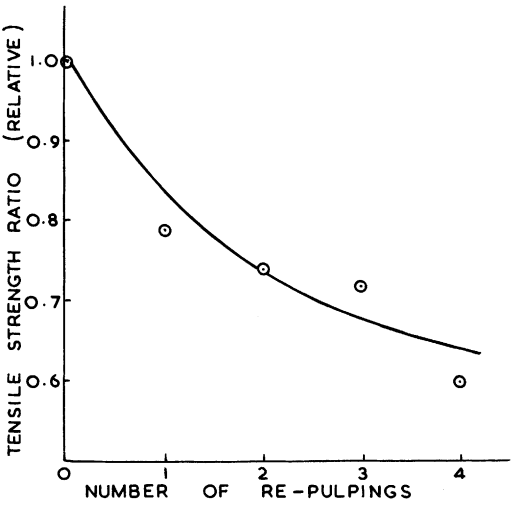

Fig. 1—Variation of tensile strength with number of re-pulpings

handsheets and then disintegrate and re-make, carrying this on for several stages; we find a reduction in tensile strength, stage by stage. The curve relating strength to number of cycles is a curve of diminishing effect, generally indicating that after six cycles or so the reduction of strength per cycle is quite small. The magnitude of the strength reduction is dependent on the type of fibre used, being smallest for mechanical pulps. The fall off in strength with successive cycles is observed when the freeness of the stock is the same at each cycle. Strength may be recovered by increased beating at the expense of increased wetness. Szwarcztajn and Przybysz[5] have shown that the major contribution to the increased wetness comes from the fine fraction. If this could be removed, beating could recover the original strength, at least partly, without the wetness increase. McKee,[6] however, reports that the maximum burst strength that could be developed, by refining five times repulped Kraft, was some 6 per cent below that of virgin stock.

The cause of the loss in tensile strength with recycling is generally accepted as being due to a loss in bonding between fibres. McKee[2] reached this conclusion from measurements of Z-direction strength and scattering coefficient and also inferred it from the observation that tear strength increased with recycling. It is well known that increase in bonding, e.g. by beating, reduces tear. Page[7] has suggested a method of separating the strength of paper into two components due to fibre and bond contributions. He proposed that, if tensile strength is regarded as a resistance to fracture, the two component resistances should be added in parallel by electrical analogy. This gives the simple equation:

$$\frac{1}{T} = \frac{1}{F} + \frac{1}{B}$$

where T is the tensile strength and F and B the fibre and bond strength components respectively. Page goes on to develop the individual terms and adduces evidence that the original hypothesis is reasonable. If we keep the equation in its simplest form and take zero span tensile measurements, with appropriate correction, as measurements of the F term; then its application to recycling results confirms that the progressive loss in strength is brought about mainly by a reduction in the strength with which fibres are bonded together. Cildir and Howarth[4] applied this analysis to recycled sulphite and Rogers and Howarth[8] applied it to a newsprint (80 per cent mechanical, 20 per cent chemical) furnish. In both cases the reduction in strength was shown to be due to loss in bonding (Fig. 2 and Fig. 3). An interesting confirmation of this comes from the, as yet, unpublished results of methods of recovering strength in recycled sheets by the addition of starch. Weight for

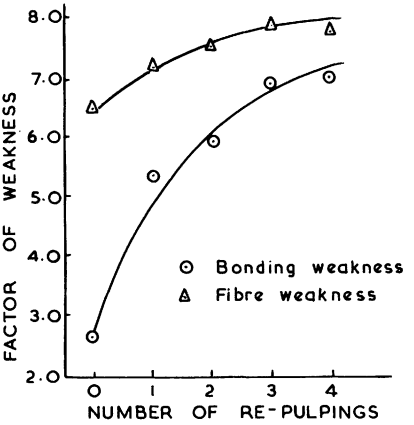

Fig. 2—Effect of the number of repulpings on bonding and fibre weaknesses, sulphite

weight retained, size press added starch is much more effective than is beater added starch. This may be explained by reasoning that the post-formation addition, on drying, causes the starch to be concentrated at fibre crossings, thus reinforcing the weakened bonds.

The classic handsheet experiment is, of course, rather artificial in that hardly ever is paper made from a furnish consisting entirely of waste paper of the same grade. Newsprint at Garden State[9] and Kent Kraft's Rhino[10] may be two exceptions. In general, we either use one grade of waste to make another grade of paper, or incorporate a proportion of waste into the furnish for the same grade. The latter is now common in newsprint manufacture.

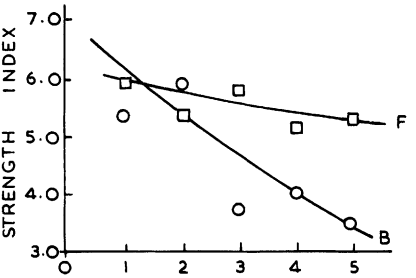

Fig. 3—Effect of the number of repulpings on bond and fibre strengths, newsprint

Where this is done we may introduce a re-use factor; defined as the proportion of re-used fibre in the furnish. In a case where this became general for the grade (for example if all newsprint manufacturers maintained a re-use factor of 0·4), a stable equilibrium would be established. It would then become possible to calculate the proportion of the furnish being recycled for the n^{th} time. This is $(1-r)r^n$, where r is the re-use factor. It follows from this that the proportion of the furnish being recycled more than n times is r^{n+1}. Thus, with a re-use factor of 0·4, the proportion of the fibre being recycled more than five times is $(0{\cdot}4)^6$, i.e. about 0·5 per cent. With a re-use factor of 0·7, the proportion going round more than five times is about 12 per cent. Tables giving the proportion of the furnish recycled for specified numbers of times at various re-use factors have been published elsewhere.[8] It is not difficult to modify the classical experiment by retaining volumes of stock from various stages and mixing in the appropriate proportions. Values of tensile, and indeed any other property, may be plotted against re-use factors. With the handsheet procedure this has been shown to give a reduction in tensile as the re-use factor is increased for sulphite[4] and newsprint.[8] The cause of this reduction is also shown to be loss in bonding, as would be expected.

Wiegand and Becker[11] performed similar experiments and showed that the effects of successive re-pulpings may be reduced by the addition of virgin pulp to the furnish. They also showed that rosin-sized paper produced a less strong sheet on recycling than unsized paper; which is in agreement with the findings of Horn.[19]

The evidence from classical, handsheet experiments seems to support the contention that paper is an inferior papermaking raw material and that its inferiority arises because of its weaker bonding potential.

Of course, the picture is not entirely bleak. I am told that a grade of paper was once advertised as superior because it had been twice made, though my informant could not give me details. Gartshore[12] reported improvements in runnability and formation obtained from the inclusion of waste in the furnish. Stockmann[13] and Rance,[13] in discussion, both called attention to the benefits obtainable by the inclusion of waste in a papermaking furnish.

The effect of papermaking on fibres

If fibres obtained from paper are inferior to fibres from pulp, in particular in their bondability, what changes in their nature are brought about by making them into paper? The bonding together of two fibres depends on two things: the area of contact between them and the formation of bonds where they touch. The former depends on the flexibility of the fibres and the latter on their surface properties. Page and Tydeman[14] reported that, on

re-wetting dried fibres, only 70 per cent of the original wet width was recovered. Jayme and Hunger[15] studied the phenomenon of hornification and noted that the fibrils are compiled into larger units (strings) on drying, thus reducing the area accessible to water and forming a stiffer structure. Page and Degrace[16] showed irreversible changes in the wall structure of beaten fibres due to drying.

In some measure, the fibre properties may be restored by treatment. This procedure is of two kinds. On the one hand, work may be done by beating or soaking in alkali[17] and these methods have a restorative effect on the fibre. On the other hand, compensating procedures may be introduced into the papermaking process to enhance the interfibre bonds. The application of starch at the size press has already been mentioned. Another interesting method is the addition of ammonium zirconium carbonate. This has been shown to improve the strength of paper made from virgin pulp[18] and an experiment at UMIST has shown that its addition on the first recycle, in a handsheet progressive recycling sequence, goes about halfway to restoring the original strength of the sheet (Fig. 4). This addition appears not to inhibit the subsequent re-pulping of the sheet, whether drying has taken place at room temperature or at 100° C. We are not yet prepared to commit ourselves to a description of the mechanism of this effect. Analysis of this and other procedures, even though they may not be commercially viable treatments, may lead us to a better understanding of the effects of papermaking on fibres. We are continuing this approach at UMIST in a study of reaction rates on fibres and hope for more results to present to the Symposium. If a good method of measuring the surface activeness of fibres emerges, it will be a useful

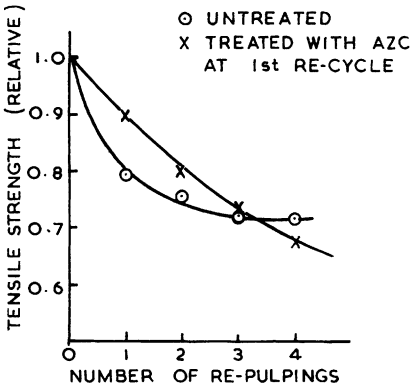

Fig. 4—Effect of AZC addition at first re-cycle on sulphite handsheets

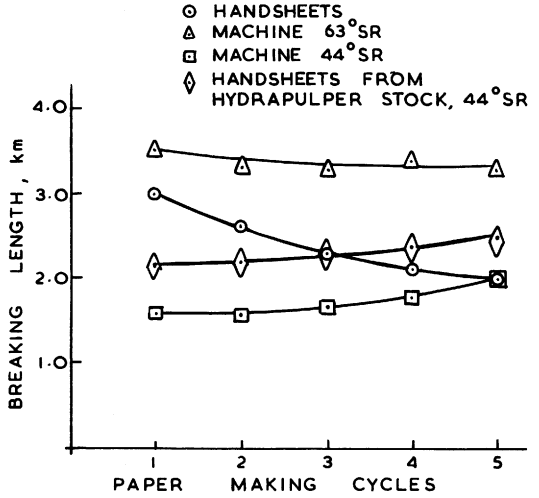

Fig. 5—Variation of breaking length with number of paper making cycles

tool in determining the effects of variations in papermaking procedure on the re-useability of fibres.

The effect of papermaking procedures

ALL the work so far discussed, and indeed nearly all the basic work on recycling, has been done with handsheets. If papermaking reduces the potential of fibres for further papermaking, it is reasonable to ask if the method of papermaking affects the magnitude of the effect. Results at UMIST reported elsewhere[8] have shown considerable differences between the effects of recycling handsheet-made paper and machine-made paper (Fig. 5). With progressive recycling we found, at one freeness, about half the strength drop with machine-made paper as compared with handsheets of the same newsprint furnish (80 per cent mechanical, 20 per cent chemical). At a higher freeness the machine-made paper showed no fall in strength with recycling; indeed a slight rise. Machine-made paper recycled as handsheets followed the same trend as the paper machine results. This indicates that, with this furnish, under the conditions obtaining in those particular experiments, it was possible to get different recycling performance due to differences in the manner in which the original paper was made. This means, in turn, that for fundamental work to have relevance to practical papermaking, it should not be confined to handsheet investigation.

Another aspect of papermaking procedure is, of course, the addition of fillers and chemicals. Horn[19] has pointed out that a rosin-sized paper will produce a less strong sheet on recycling, because of the reduced wettability of the fibres. Klungness[20] performed a series of experiments by recycling paper contaminated by various common additives such as ink, wet strength resin and polyethylene. The contaminants were removed in the recycling process. By running an overall control using uncontaminated and untreated material, and also putting uncontaminated material through the removal treatments, he was able to separate the effects of contamination and removal treatment. His results indicate that the removal is of greater effect than the contamination and that de-inking nearly restored the original strength of the sheet; presumably because of the effect of the caustic used.

Conclusion

IT is suggested that the fundamental problem in recycling is the effect of papermaking on fibres. The act of making them into paper conditions them for their subsequent use. Indeed, de Ruvo and Lundberg[21] have suggested that fibres have a long enough 'memory' for the original pulping process to affect their ultimate potential for use as secondary fibre.

In order to measure the essential difference between secondary and virgin cellulose fibres we need a technique to measure their surface properties, chemical and physical. Armed with such a technique, we could then investigate the relationships between papermaking parameters and the quality of secondary fibres generated when paper becomes waste paper. The search for such a technique is currently occupying us at UMIST. Because a sheet of A4 paper contains 6 million fibres, techniques which look at fibres half a dozen at a time may be difficult to apply.

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Discussion

Prof. E. Szwarcstajn Following that very good review of the state of knowledge in the field of recycling, I would like to add that, in an attempt to find new methods which could allow improved mechanical properties of the paper from secondary pulps, without encountering the usual drainage problem, new ideas have been proposed. They include, among others: flushing the pulp in an alkaline medium; a very mild refining; a beating at high consistency; fines separation; addition of various chemicals and so on. But now I feel that to find a really effective method of regeneration of the paper making capacity of recycled pulps, we must have a better understanding of what is occurring in the recirculated pulp—that is, in its fibres and in its fines. What changes does it undergo during refining, forming and drying, which are the main cause of its loss of bonding capacity? This problem is not just purely technical, but becomes more and more scientific and fundamental. I think that it was a good idea of the committee of this symposium to include the topic of recycling in the programme because it allows us to discuss with such a distinguished audience: 'Where are we now and are we going the right way?'

Dr A. de Ruvo There is a discrepancy between different countries concerning their Recycling Factors. Sweden has a rather low one whilst, maybe, Holland and Germany have high ones. Referring to your curves and taking, for example, the case of liner board—when is there a critical point where the deliveries of virgin pulp or virgin fibres must be kept to certain limits in order to keep the properties of the European test liners to certain levels? Can you expand a little on that problem which is, I think, important for both of us.

Howarth Yes, that is the same point I was making. There must be some level of re-use below which we cannot go without going out of specification for quality. I think that is the point you are making, isn't it? The point I was trying to make was that, if you want to find that level, then you must do your experiments and plot your graphs with the Re-use factor as the abscissa and not the number of cycles. You will not find that point by doing the

classic experiment. You will only find that point if you do your experiments from a Re-use factor point of view.

Mr D. Attwood It is said that within every fat man there is a thin man struggling to get out. It must be true that within every wood fibre, or any other fibre for that matter, is a papermaking fibre struggling for release. If the pulp makers are clever, and the fibre is released in the right way, we have the perfect fibre for its particular papermaking role. All that is necessary is to take away those layers of the original fibre we do not want and, with a mechanical fibre, we are prepared to accept almost the whole wood fibre with all its deficiencies in exchange for a virtual 100 per cent yield. We will pay the price of a 60 per cent yield pulp because we like the properties of that particular pulp. Of course we recognise that yield is not a single parameter. Different cooking processes will remove different constituents of the original fibre and the problem facing those of us working on recycled fibres if, firstly, that the original work carried out to release the papermaking fibre was not always perfect and, secondly, and much more importantly, that irreversible changes have taken place within the fibre during the papermaking process. One of the consequences I find of speaking on the last day of this conference has been to bring home to me the looseness of our phrase 'virgin fibre' and I should immediately explain in reference to the PIRA paper that we have used 'virgin' to mean 'that pulp never used to form a sheet of paper'. However, in listening to, for example, Dr Scallan, it is immediately obvious that a distinction needs to be drawn between never dried and once dried fibres. The virgin fibres of a U.K. paper maker derive in the main from baled pulp. I am now considerably concerned that, what we call the first recycle, is in fact, the second. Regretfully, the situation is not as clear as that, because, obviously, in the case of pulp manufacture, little or no refining has been attempted. The manufacturers hope that our reslushed pulp is close in properties to the original product. It would have been nice to have found an alternative to the expression 'virgin fibre', which to me seems to permit of only two states, 'either/or' and I thought perhaps Stratford on Wednesday night might have provided a sufficiently earthy alternative, but Shakespeare seemed able to distinguish between 'virgins' and 'spotless virgins' and I realise that experimental technique alone would be unable to distinguish the two.