

# ***Improvements in Strength of Polymer-Containing Cellulosic Networks by Heat Consolidation***

## **INTRODUCTION**

Networks of cellulosic fibers formed in absence of water have a structure and a mechanical property profile different from those of conventional wet-laid paper products. A dry-formed cellulosic network (or dry-formed paper) can be said to be a product somewhere between ordinary paper and a nonwoven textile. There are several reasons for the recent interest in dry-formed paper structures, of which the water absorbancy and softness of such structures can be mentioned, together with the possibility of easily forming structures from mixtures of different fibers and also some advantages relating to the process technology itself.<sup>1,2</sup>

The strength of dry-formed cellulosic networks is initially low, only about one tenth of the strength of wet-laid paper at a comparable density.<sup>2</sup> A strength enhancement of the loosely bonded fiber structure is thus required. The low tensile strength of the network is due primarily to the lack of sufficiently strong fiber-to-fiber bonds, i.e., the stress transfer between crossing fibers is poor. In practice, the fiber bonds are strengthened by the use of synthetic polymeric binding materials.<sup>3,4</sup> Normally a latex is sprayed to both sides of the fiber web and a dry-formed paper of sufficient strength usually contains 10–20% by weight of a synthetic polymer, e.g., an acrylate copolymer.

In an earlier work<sup>4</sup> it was shown that the tensile strength of dry-formed paper may be made several times greater by choosing a suitable polymeric binder. In that case, the networks were impregnated with a solution of the polymer or with a polymer dispersion of low solids content and then allowed to air-dry at room temperature without restraint. In the present communication, it is shown that if pressure is applied and the temperature increased during drying of the network, the strength (and stiffness) of the bonded networks is further increased. It is shown that this improvement is due not only to the enhanced softening and flow of the polymeric binder but also to the plasticization of the wood polymers constituting the fibers induced by a combination of water and temperature. The binder content is here kept below 10% by weight.

## **MATERIALS**

The dry-formed web was produced from a commercial defibrated grade of fluff pulp (bleached sulfate). The basis weight of the networks was ca. 90 g/m<sup>2</sup>. The synthetic binder was based on a copolymer of butyl acrylate and acrylonitrile (AC) with a glass transition temperature of 12°C and a density of 1.1 g/cm<sup>3</sup>. It was added to the network in the form of a water-based latex.

## **EXPERIMENTAL**

The dry-formed sheets were made as described earlier<sup>4</sup> and were immersed in a dispersion of an appropriate solids content. They were then dried for 15 min in a press at four different pressure levels (3.3, 10, 15, or 20 MPa) and at three temperatures (50, 80, or 105°C). Prior to pressing, the water content of the sheets was ca. 80%. For comparison a series of dry-formed sheets which were immersed only in water were subjected to the same drying conditions. After the pressing stage the sheets were allowed to dry in air (room temperature) without restraint.

The mechanical properties of the bonded networks were evaluated at 23°C and 50% relative humidity (RH). The samples were conditioned at 23°C and 50% RH, prior to being tested. The conditioned test specimens contained ca. 7–8% water.

## RESULTS AND DISCUSSION

Before reporting on the mechanical properties of the bonded sheets, it should be mentioned that the scatter in material properties is in general pronounced for dry-formed networks. This reflects only the inhomogeneous structure of such materials.<sup>4</sup>

As already mentioned, the sheets contained ca. 80% water prior to pressing. After 15 min at an elevated temperature and at a higher pressure the water content had decreased. At the maximum pressure used here (20 MPa) the moisture content had decreased to ca. 60% at 50°C, to ca. 50% at 80°C, and to ca. 30% at 105°C. The pressure level had only a minor effect on these moisture contents. A decrease in pressure to 3.3 MPa resulted in ca. 5% higher moisture content at each temperature level. After being pressed, the sheets were allowed to air-dry without restraint. This results in lower strength and lower stiffness of the bonded structure than in sheets restrained (pressed) until they are almost completely dry.<sup>5</sup> This effect is more pronounced for sheets subjected to the lowest temperature, i.e., 50°C, due their rather high moisture contents after the pressing operation.

Due to the relatively high water contents and to the fact that part of the drying was carried out without restraint, the increases in network density were rather modest. Prior to the pressing the density of the dry-formed sheets was ca. 370 kg/m<sup>3</sup>. At the highest temperature 105°C, the increase in density was approximately 110 kg/m<sup>3</sup>, and at lower temperatures significantly less, 50 kg/m<sup>3</sup> or below. These figures depend somewhat on the polymer content, but can be taken as rough guidelines in this context.

Having discussed briefly the effect of pressing on the structure of the networks, attention now can be focused on the influence of the binder content on the strength characteristics. Figure 1 shows the specific strength, i.e., the tensile strength divided by the bulk density, vs. the content of the acrylate copolymer for a sheet pressed at 50°C and a pressure of 20 MPa. Only small amounts of the polymeric binder are evidently required to raise the specific strength appreciably. The strength increases linearly with the amount of binder in this range of binder content. The behavior depicted in Figure 1 is typical for all combinations of pressures and temperatures used in this study and is also to be expected from the results reported earlier.<sup>4</sup>

Figure 2 is a summary of all the results regarding the specific strength ( $\sigma_B/\rho$ ) of the bonded dry-formed networks. In this graph all strength values have been normalized to a constant binder content level of 2.5%.

Values of the specific strength of dry-formed sheets which have been immersed in water only are also included in this figure. Several features of Figure 2 are noteworthy. Firstly, it should be pointed out that, with a suitable combination of temperature and pressure, sheets having specific strengths of the order of 10 N m/g can be obtained even at this low binder content. A dry-formed sheet, with 2.5% of this binder, which has been allowed to air-dry without restraint has a specific strength of only ca. 1 Nm/g.

An increase in temperature and/or pressure clearly improves the strength values markedly. It is, however, particularly interesting to note that the moisture itself has a significant effect

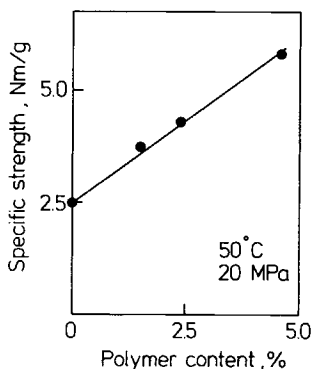


Fig. 1. The specific strength ( $\sigma_B/\rho$ ) of the bonded dry-formed networks vs. the amount of polymeric binder (acrylate copolymer). The pressure and temperature during the press stage were 20 MPa and 50°C, respectively.

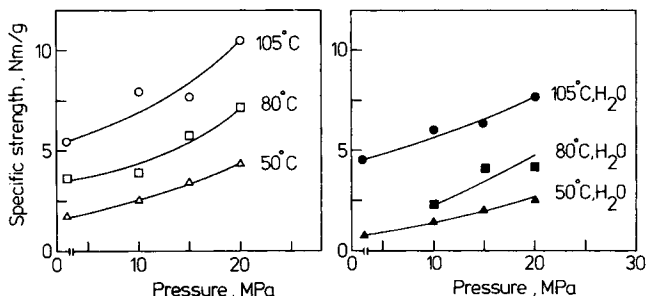


Fig. 2. The specific strength ( $\sigma_B/\rho$ ) of the dry-formed networks vs. the applied pressure at different temperatures. The unfilled symbols denoted values for sheets containing 2.5% acrylate copolymer and the filled symbols refer to sheets which have been immersed only in water. Key for temperatures ( $^{\circ}\text{C}$ ): ( $\blacktriangle, \triangle$ ) 0; ( $\blacksquare, \square$ ) 80; ( $\bullet, \circ$ ) 105.

on the strength values. The networks which have been immersed in water only show a strength increase with temperature and pressure similar to that in the polymer-containing sheets. Water alone can thus improve the bonding state of the initially rather weak fibrous structures. This is in agreement with the results reported by Byrd<sup>6</sup> and by Back et al.,<sup>7</sup> who used only moisture to improve the mechanical properties of dry-formed paper. The contribution to the strength values by the polymer can be estimated by subtracting the strength values of the "water-immersed sheets" from the corresponding values for the polymer-bonded networks. This contribution is of the order 1–2 N m/g at a polymer content of 2.5% and it is not very sensitive to changes in pressing conditions or temperature, although an increase with pressure can be noted at 105°C.

To a first approximation it appears that the improvement in strength of the networks under the action of pressure and temperature is associated with the moisture, while the polymer contributes an approximately constant amount determined by the amount of the binder in the sheets. It should be pointed out that this behavior is probably related to the fact that the binder used here has a low glass transition temperature ( $T_g$ ) and readily forms a film already at room temperature. Further increases in temperature and pressure do not apparently further improve the effect or the binder distribution in the network. If, however, a binder with a high glass transition temperature is used, e.g., polystyrene, an effect of temperature, on the polymer contribution to the specific strength of the dry-formed network is certainly to be expected when it is increased through the  $T_g$  region.

In the present case the improvement in strength when the temperature and pressure are increased is likely to be primarily due to plasticization of the wood polymers (hemicellulose and less ordered regions of cellulose) due to the combined action of water and temperature<sup>8</sup> during consolidation of the structure.

For reasons of completeness, it should be mentioned that for sheets pressed at 80 and 105°C the elongation at rupture is rather low, of the order 2–3%. For sheets pressed at 50°C, the ductility is somewhat higher, especially for structures compacted at lower pressures, for which values up to ca. 7–8% can be obtained. This is partly due to the fact that such sheets have the highest moisture content after the pressing stage and are thus not restrained for a major part of the drying period (cf. also Ref. 5).

#### FINAL COMMENTS

It has here been shown that the strength of polymer-bonded dry-formed paper sheets can be increased considerably by applying pressure and increasing the temperature. It is obvious that the mechanical properties of networks of this kind be controlled by choosing the processing conditions. In addition to the density change, there are at least three factors noted here that can influence the strength of networks which have been impregnated with a latex. The amount and type of synthetic polymer-based binder is of course important.<sup>4</sup> The combination of the softening of the wood polymers by water and high temperature and the compacting of the structure also increase the strength of fiber—fiber bonds and the number of such bonds. The

drying strategy is also significant in this case. If the sheet is restrained during drying to a low moisture content, an improvement in strength is obtained.<sup>5</sup>

In networks dried in the way employed in this work, there will, in a simplified sense, be two types of bonds between crossing fibers; fiber—fiber bonds rather similar to those prevailing in wet-laid paper and fiber—polymer—fiber joints. From the results presented here it appears that the contributions from the polymeric binder and from the moisture-induced softening of the wood polymers are additive. On the fiber level the situation is slightly more complicated, since some of fiber surfaces that take part in bonding when only moisture is used to improve the strength are obviously covered with the polymeric binder when the network is immersed in the polymer dispersion. A more detailed study of possible changes in binder distribution due to increases in temperature and pressure would be of value in this context. Such measurements are, however, difficult to perform and are beyond the scope of this short communication.

Finally, it should be mentioned that it is naturally possible to improve the mechanical data reported here, e.g., by choosing another processing time and by choosing other temperature and pressure levels. In practice, one would certainly work with significantly shorter press times and probably combine this with an increase in temperature. The aim of this work has not been to produce very strong structures, but merely to clarify the importance of the processing conditions in relation to the mechanical properties of polymer-bonded dry-formed networks of the cellulosic fibers.

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### References

1. B. W. Attwood and D. G. White, *Tappi*, **62**(2), 39 (1979).
2. A. de Ruvo, H. Hollmark, S. Hartog, and C. Fellers, *Sven. Papperstidn.*, **85**(7), 16 (1982).
3. J. A. Villalobos, *Tappi*, **64**(9), 129 (1981).
4. M. Rigdahl, B. Westerlind, H. Hollmark, and A. de Ruvo, *J. Appl. Polym. Sci.*, **28**, 1599 (1983).
5. M. Htun, Ph.D. thesis, Royal Institute of Technology, Stockholm, Sweden, 1980.
6. V. L. Byrd, *Tappi*, **65**(5), 153 (1982).
7. E. L. Back, S. C. G. Wahlström, and R. G. Andersson, *Tappi*, **62**(3), 85 (1979).
8. L. Salmén, Ph.D. thesis, Royal Institute of Technology, Stockholm, Sweden, 1982.

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