

Bond setting in the corrugating operation

The rate of dehydration of the starch adhesive

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The mechanism of dehydration is predominantly one of molecular diffusion of water into fiber walls. Factors that slow down the transport of water to the sorption sites, such as sizing, decrease dehydration rate.

In the setting of starch adhesive on paperboard at the corrugator, heat causes the raw starch granules in the starch solution to melt into a gelatin, and the paste viscosity increases dramatically. This unique property of starch is responsible for the development of initial bond strength. Concurrent with this process (1, 2), the aqueous adhesive is dehydrated by both the corrugating medium and linerboard, which absorb water. The cohesive strength of the starch film is raised further, until the adhesive layer is completely dried.

Paperboard may absorb water in at least two ways:

1. By capillary absorption in the interfiber pore network
2. By diffusion of water molecules into fiber walls leading to swelling.

Both mechanisms may be operating concurrently (3). The penetration of water into paper and linerboard by capillary action has been studied in recent years (3, 4). The progress of water penetration is dependent on the square root of time, suggesting that the paper can be treated as a series of capillaries. Washburn's equation (5) is often used, although its applicability at short times of contact has been questioned (6). In contrast, little has been published on the kinetics of water vapor diffusion into paperboard except for a few studies done with regenerated cellulose (7-9).

The full thermal gelatinization of starch, which occurs by heat transfer from the heated linerboard, is not instantaneous (10). There is a short period during which the aqueous phase in the glue could be removed by a combination of capillary and hydrodynamic pressure, leaving the uncooked starch granules (10-50 μm diameter) filtered off at the surface pores (about 5 μm). Since the cooked starch concentration is only about 3%, the migration of the aqueous phase into the linerboard would raise the overall concentration of the remaining layer, increasing

its viscosity and "tack." As the starch granules swell and gelatinize, less and less free water is available, and the solution viscosity increases sharply.

Thus, the dehydration at this stage should be mostly through water vapor diffusion. The dehydration mechanism can therefore be expected to change from (a) a combination of both capillary penetration of water in the aqueous phase and water vapor diffusion, at short times, to (b) predominantly water vapor diffusion during and after gelatinization, at relatively longer times. The relative contribution of each mechanism to the rate of dehydration of starch adhesive remains unknown.

Here, we first examine how the dehydration rate of the adhesive is affected by various factors such as sheet structure, basis weight, sizing, and fiber type. Then, by examining how these affect capillary absorption and the molecular diffusion of water, conclusions are drawn regarding the mechanism of adhesive dehydration.

Experimental procedures

Preparing and characterizing starch adhesive

A typical "carrier type" starch adhesive was prepared according to a recipe described by Kroeschell (11). Total starch concentration was 20.5% by weight, of which the cooked portion was 2.9%. The paste viscosity was 0.53 Pa·s at 22°C, as determined in a Brookfield viscometer. Its surface tension was 51 mN/m, as determined by the pendant drop method.

Preparing and characterizing the paperboards

Paperboard handsheets were made from unbleached dried kraft pulp at two levels of beating, 560 mL and 408 mL CSF. The basis weight of the sheets was varied from 75 g/m² to 300 g/m². The sheet density was changed by wet pressing or beating. The porosity and pore size distribution of the linerboards were determined by mercury porosimetry. One sample of 150 g/m² was sized with alkyl ketene dimer (Aquapel 365, Hercules) in hexane solution using the procedure described by Huynh *et al.* (12). Table I summarizes the characteristics of the linerboards. Several

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I. The effect of wet pressing and beating on sheet properties

Basis weight, g/m ²	Wet pressing pressure, kPa	Caliper, μ m	Air permeability, mL/min	Parker Print-Surf, μ m	Density, kg/m ³	Pore size, μ m
560 mL CSF						
75	35	237	2270	6.6	316	...
	350	139	2000	5.6	540	...
	3500	108	752	3.4	694	...
150	35	412	2160	7.2	364	12.5
	350	278	1740	6.1	540	7.0
	3500	185	347	3.9	810	3.5
225	35	683	2050	7.4	329	...
	350	429	1490	6.5	524	...
	3500	285	196	4.5	789	...
300	35	744	2030	7.3	403	...
	350	583	1370	6.3	515	...
	3500	378	172	4.3	794	...
408 mL CSF						
75	35	145	531	6.7	517	...
	350	109	262	4.7	688	...
	3500	95	46	2.8	789	...
150	35	277	343	7.6	542	6.5
	350	203	86	5.2	739	3.5
	3500	171	16	3.9	877	1.7
225	35	398	257	7.7	565	...
	350	319	62	5.9	705	...
	3500	266	16	3.5	846	...
300	35*	608	264	8.1	493	...
	350	398	43	6.6	754	...
	3500	346	11	4.5	867	...

commercial paperboards of the same basis weight were used to test the effect of pulp type on the rate of moisture absorption.

Measuring water absorption

Three types of measurements were made:

1. Rate of dehydration of adhesive
2. Absorption rate with the liquid water absorption tester
3. Vapor sorption rate.

The dehydration rate of the adhesive placed in contact with the paperboard sample was followed by monitoring the change in infrared absorption with time using multiple internal reflectance (MIR), according to a technique described elsewhere (12). Infrared (IR) light of wave-number 3400 cm⁻¹ is strongly absorbed by water. As water leaves the adhesive, the absorption decreases (or the reflectance increases). The increase in reflectance with time is therefore a measure of the increase in concentration of the adhesive.

The short time absorption of liquid water by the paperboard was measured using the Bristow absorption

tester (13). Both MIR and short time absorption measurements were carried out in a controlled room at 22°C and 50% RH.

Measurements of water vapor absorption were made with an electrobalance connected to a sorption chamber. First the chamber is evacuated. After subjecting the sample to a vacuum of 0.6 kPa, water vapor was introduced, causing a sudden change in relative humidity from 0% to 50%. The weight increase of the sample was continuously recorded. The experiment was repeated, raising the relative humidity from 50% to 85%.

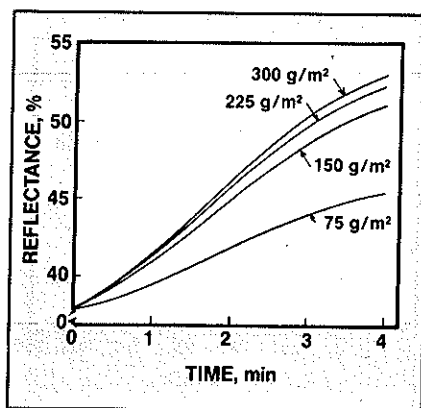
Results

Adhesive dehydration

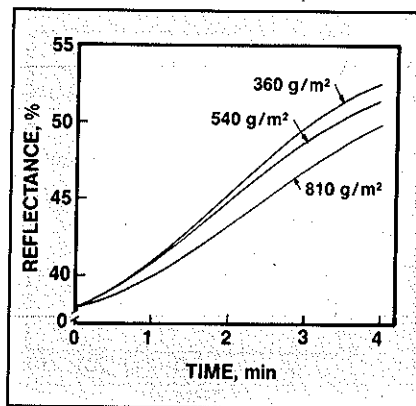
Figure 1 shows the effect of sheet basis weight on the dehydration rate. The initial slope of the curve is considered to be a measure of the dehydration rate. Above 150 g/m², basis weight has little effect on adhesive dehydration. Decreasing the basis weight to 75 g/m², however, considerably reduced the rate.

Increasing the sheet density by increasing wet-pressing or beating decreased the dehydration rate (Figs. 2 and

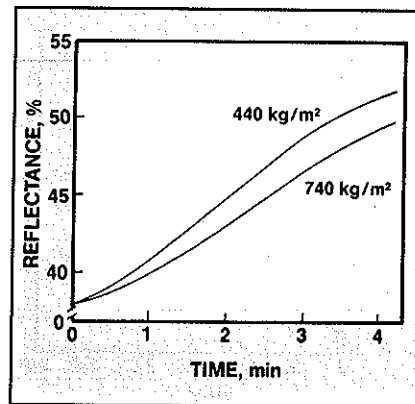
1. Dehydration of starch adhesive by paperboard handsheets of increasing basis weight



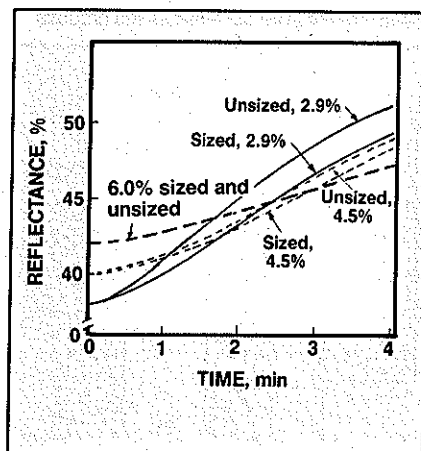
2. The effect of sheet density, varied by wet-pressing, on the rate of dehydration of the adhesive



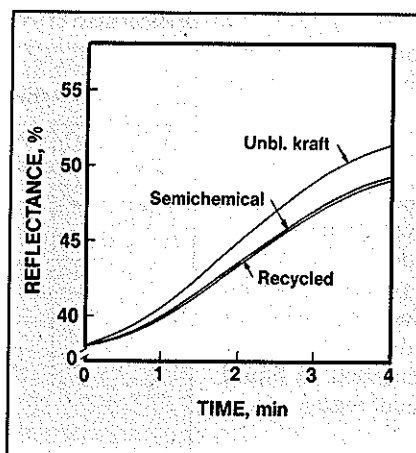
3. The effect of sheet density, varied by beating, on the rate of dehydration of the adhesive



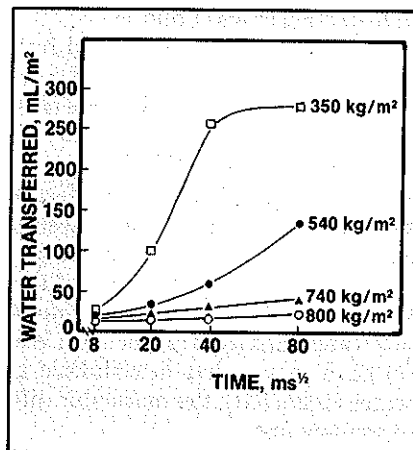
4. The effect of sizing on dehydration rate (The initial value of reflectance decreases as the water concentration in the adhesive increases.)



5. The effect of pulp type on the rate of dehydration of starch adhesive



6. The effect of sheet density on the rate of water absorption measured by the Bristow tester



3), the effect being most notable above a density of 540 kg/m². The air permeability changes significantly in this range of density (Table I).

We examined the effect of sizing with adhesives formulated with the same concentration of raw starch (20.5%) but increasing concentrations of dissolved cooked starch. We increased the dissolved cooked starch to increase the viscosity of the aqueous phase. Figure 4 shows that at a cooked starch concentration of 2.9%, sizing affects the dehydration rate. As the concentration increased to 4.5%, the effect was reduced until, at 6%, no difference was observed between sized and unsized sheets. Note that the difference between sized and unsized was slight relative to the overall change in reflectance, suggesting that capillary phenomena do not play a major role in the dehydration process.

Figure 5 shows the influence of the type of fiber. Kraft fibers are more efficient than recycled and semichemical fibers in dehydrating the adhesive.

Capillary absorption and diffusion of water

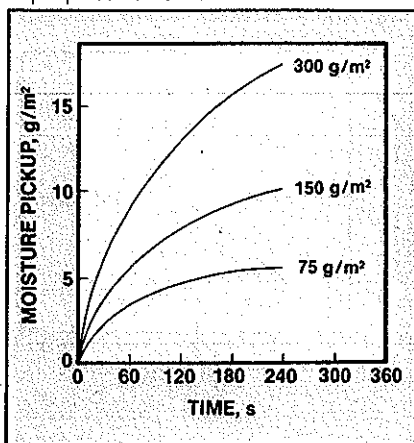
While no measurements were performed, there is no reason to suspect that the initial rate of capillary

absorption of water would be significantly affected by the sheet basis weight. Sheet density had a considerable effect on the capillary absorption of liquid water, as shown in Fig. 6. The Washburn equation predicts a decrease in the rate with a decrease in capillary diameter. The Washburn equation describes capillary flow into cylindrical pores as induced by surface tension.

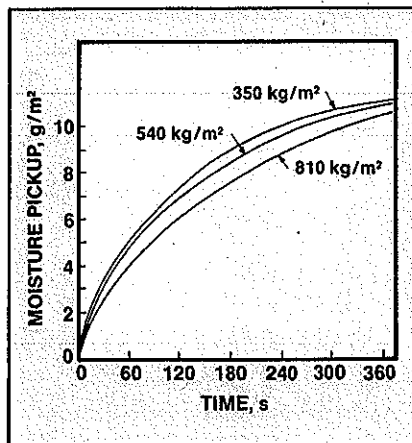
Figure 7 compares paperboard samples of increasing basis weight for moisture pickup, or the diffusion of water vapor into the sample. The overall absorption process results from the diffusion of water vapor through the porous network of the sheet, with concurrent intrafiber molecular diffusion. The basis weight influences the diffusion rate.

The amount of water absorbed into the 150- or 300-g/m² sheets is less than twice or four times the amount absorbed into the 75-g/m² handsheet. This difference is explained by considering that the fiber surfaces inside the sheet are not in contact with the same vapor pressure as the outside fibers (14). In other words, pore diffusion can become controlling as the diffusion distance increases (i.e., increasing sheet basis weight) or as the pore size decreases. Figure 8 confirms this view, showing that diffusion slowed

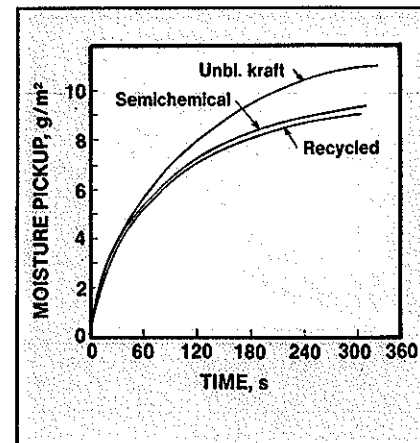
7. The diffusion of water vapor into sheets of increasing basis weight after a step change in vapor pressure from 0% to 50% RH



8. The effect of sheet density on the rate of moisture absorption after a step change from 0% to 50% RH



10. The effect of pulp type on the rate of moisture regain



as sheet density increased. Note the similarity in the effect of basis weight on vapor diffusion and on adhesive dehydration (Figs. 1 and 7).

Sizing, when it is performed in the manner described here, does not affect water vapor sorption but completely suppresses capillary absorption (12).

Mechanism of dehydration

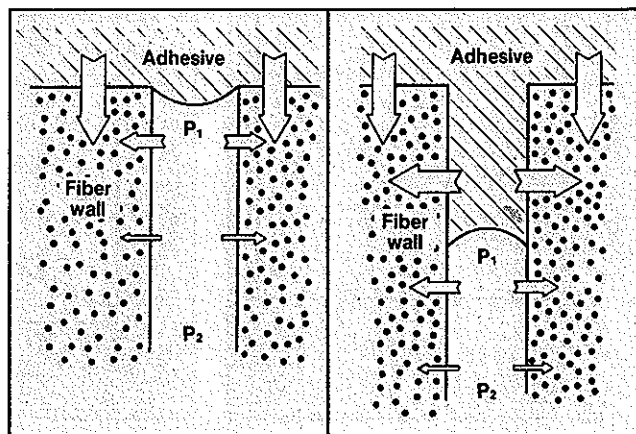
The similarity between the effect of sheet basis weight, sheet density, and sizing on adhesive dehydration rate and on water vapor diffusion is clear. No doubt, intrafiber diffusion is an important part of the dehydration process. While the diffusion of water into cellulosic materials at low water activity is rather slow, Westman and Lindstrom (9) have shown that in cellulosic gels exposed to liquid water (100% RH), the molecular diffusion of water can be extremely fast.

The second process, capillary flow, may also play a role in the early stages. Capillary movement of the aqueous phase could help increase the solids content of adhesive in two ways. First, as mentioned earlier, one may consider that the absorption of the liquid phase with filtration of the starch granules would automatically result in an increase in the concentration of the unabsorbed top layer (probed by the infrared radiation).

Second, by penetrating into the sheet, the aqueous phase (100% RH) would be in direct contact with a large number of fiber surfaces, as sketched in Fig. 9. The vapor pressure gradient ($P_1 > P_2$) causes water vapor diffusion along the pore. Intrafiber diffusion takes place concurrently (arrows). The penetration of the adhesive into the pore accelerates the process. Without this capillary flow, these surfaces would be in contact with water vapor of lower activity if a gradient in vapor pressure exists along the pores; hence, the force impelling diffusion would be lower. (At the corrugator, capillary flow into both linerboard and medium would be expected to be caused predominantly by the hydrodynamic pressure in the corrugating nip, although flow driven by surface tension cannot be ruled out in the unsized medium.)

The faster dehydration rate observed on the unsized sheet with the adhesive of lower viscosity is believed to be caused by the effect of increased interfacial contact.

9. A: The vapor pressure gradient ($P_1 > P_2$) causes water vapor diffusion along the pore. Intrafiber diffusion takes place concurrently (arrows). B: Penetration of the adhesive into the pore accelerates the process



Finally, the ranking of the three pulps in terms of water vapor sorption (kraft, recycled, and semi-chemical) in Fig. 10 is the same as that observed in the adhesive dehydration experiment of Fig. 5.

Conclusions

Overall, we conclude that the mechanism of dehydration is predominantly one of diffusion of water from the adhesive into the fiber walls. Factors that restrict the transport of water molecules to the sorption sites—such as sizing and increasing density—decrease the dehydration rate. On the other hand, factors that reduce the force that causes diffusion—the initial moisture content of the sheet—would be expected to slow down dehydration. Fiber type also affects diffusion rate. □

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Received for review March 15, 1989.

Accepted Aug. 10, 1989.

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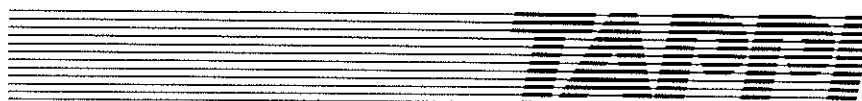
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