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ANISOTROPY OF INTERNAL STRESS RELATED TO PAPER SURFACE ROUGHENING

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ABSTRACT

Release of internal stress has been referred to as one of the causes for surface roughening of paper. However, occasionally drying-induced stress is not corresponding to the roughening of the sheet. The authors assumed that roughening is not caused by release of in-plane internal stress built in drying, but of transverse internal stress built in transverse processes such as calendering and wet-pressing. The analytical procedure of Kubát for determining in-plane internal stress was attempted to apply to the transverse compressive mode to determine the transverse internal stress. A straight-line portion was found at higher initial stresses in the plots of relaxation speed per unit logarithmic time versus initial stress (Kubát's plot). The slope and the offset of this straight-line portion were varied with the forming process the sheet subjected. The initial stress at a relaxation speed of 25 N/*log(s)* was defined as an index of transverse internal stress. Consequently, the greater the calendering, wet-pressing and beating, the higher the transverse internal stress (the slower the relaxation). But, the amount of loaded calcium carbonate did not affect it. The transverse internal stress had influence on surface roughening of paper, but the relationship was observed to be different between the papermaking processes. Besides, the transverse internal stress was likely to be associated with apparent sheet density depending on the fiber source and the way of sheet making. The authors considered that the degree of fiber collapse and viscoelasticity are closely related to the transverse internal stress.

INTRODUCTION

Surface roughening of paper in water-based processes like pigment coating and off-set printing has been considered to occur due to fiber rising and puffing[1], recovery (ribbon-to-tube) of fiber shape, interfiber debonding, fiber swelling and relaxation of internal stress[2]. Internal stress is generally defined as stress built in fibers or the fiber network during restrained drying and remaining there even after removal of the drying tension. That's why it is also termed dried-in stress, built-in stress, frozen-in stress and residual stress. Residual stress is generally conceived as being within elastic behaviors. Sasaki *et al* [3, 4] calculated and measured a residual stress distribution in curved laminated wood beams within the elasticity limit. But, he suggested the deviation of the observed stress from the calculated ones seemed to be caused by the relaxation of the residual stresses. Johanson and Kubát *et al* [5, 6] proposed a method to measure internal stress in paper by stress relaxation measurements (details will be discussed later) and showed that the level of internal stress was varied with drying tension and that it relaxed by exposing paper to water or high humidity. Without tension, paper shrinks during drying. Drying shrinkage of paper, a contrary concept of internal stress, affects its physical properties as suggested by Kimura[7]. He reported that, in tensile test, paper elongated until failure by a length (additionally to that of the unshrunk sheet) almost equal to the length the paper shrank during drying. Where and how is internal stress built in paper? Microcompression[8] at fiber crossings probably play a roll. Fiber twisting[9] caused by hydrogen bondings between fibrils running at an angle may also contribute.

However, it is doubtful that such internal stress due to drying tension is directly related to surface roughening. Additon[10] showed that exposure to 97 % relative humidity for one week hardly changed surface properties of handsheets from never-dried hardwood kraft pulp; gloss seemed to decrease moderately, but it was not significant; PPS roughness did not change (thickness increased by up to 2 %); internal stress measured according to Kubát's method decreased by 24 to 38 % depending on the beating degree. The author obtained a similar results for commercial bond paper; as a result of the same exposure, it did not show any significant change in gloss or PPS roughness, but a large decrease in internal stress of 33 %. Those are examples of internal stress reduction without surface roughening. Sasaki *et al* [11] showed that soaking commercial fine paper in water and redrying it reduced its smoothness as internal stress decreased. When the water-treated paper was calendered to an equivalent level to that of the original paper and subjected to the same water treatment again, smoothness was reduced as much as was that of the original paper while internal stress was unchanged. This is an example of surface roughening without internal stress reduction.

Internal stress mentioned so far is all connected to in-plane tension during drying. Internal stress must be anisotropic because the inducing stress applied to the material is anisotropic. So, it is natural to think that compressive force, as in calendering, likewise should build internal stress in the transverse direction of paper. Engström suggested that precalendering compressed fiber flocs in the basepaper and they expanded on coating with the larger coat weight variance, implying the more roughening than for uncalendering. To measure transverse internal stress, Kubát's method was attempted in the compressive mode. Differences found in stress relaxation curves in the compressive mode between differently processed sheets were interpreted and discussed.

THEORY

In-plane internal stress (Kubát’s method)

Kubát *et al* [5] proposed a unique method to determine in-plane internal stress based on the hypothesis of stress-aided thermal activation[12]. According to it, in-plane internal stress is determined as follows: For stress relaxation curves obtained for various target stresses in a tensile mode, the rate of stress relaxation to logarithmic time after the target stress was reached are calculated for each curve; plots between the relaxation speed and the initial (target) stress usually gives two rectilinear portions; The intercept of a rectilinear relationship at higher initial stresses in extension on the initial stress axis represents the in-plane internal stress of the specimen. Figure 1 illustrates these calculation processes.

Transverse internal stress

The same procedure was utilized for the compressive mode, empirically. Details will be explained in Results and Discussion.

Transverse internal stress seems to be generally connected with fiber collapse. Robertson[13] stated that fiber collapse is produced by wet-pressing and that frozen-in stress maintaining the fiber collapse is developed during drying. Reswelling of the dried fibers is dependent on the frozen-in stress in the way that bonding within the cell wall is dissolved wherever forces tending to restore the original shape of the fiber assert themselves. The recovery, however, is incomplete if the frozen-in stress have relaxed by time, temperature and plasticization. Here, it should be noted that compressive stress relaxation in the compressive mode is different from frozen-in stress relaxation mentioned by Robertson.

Forseth *et al*[14] showed that mechanical pulp fibers that collapsed in calendering regain their tubular shape when moisture releases the pressure-induced stresses. The author revealed that the fiber shape recovery affected surface roughening and that the effect increased with cell wall thickness.

Enomae *et al*[15,16] examined freeze-dried kraft fibers from a handsheet after soaking in water, after defibering, after beating and compared to fibers from a reslushed lap pulp and the never-dried pulp. There were no mechanical actions that perfectly recovered the tube-like shape, but only beating carried out to the reslushed lap pulp did for a very small proportion of the fibers. The work concluded that in the case of kraft fibers, shape recovery is not a significant contribution to the roughening process.

EXPERIMENTAL

Samples

Tables 1 and 2 summarize attributes of the samples used to examine effects of calendering, wet-pressing, beating and filler loading. For calendering effects, machine-made wood-containing paper consisting of a 15/40/45 ratio of softwood kraft/GWD/TMP was used. The paper with a moisture content of ca. 8 % was supercalendered off-machine on a laboratory scale at two levels of linear pressure, 29 and 49 N/m. For wet-pressing, beating and filler (calcium carbonate) loading effects, handsheets were prepared from bleached hardwood kraft pulp. For the beating series, the freeness was 639, 460 and 260 ml CSF for unbeaten, 5000 and 20000 revolutions, respectively. Neither filler nor chemicals was added to the furnish of the wet-pressing or beating series. The pulp for filler loading was beaten for 5000 revolutions in PFI mill. Calcium carbonate used was PC (Shiraishi Kogyo Kaisha. ltd., Hyogo, Japan). All the handsheets were prepared according to TAPPI test method unless stated in the table.

Smoothness

Smoothness was measured using Oken type smoothness tester (Asahi Seiko co. ltd., Chiba, Japan). As an axis title, "Bekk smoothness" was used because the principle is identical to that of Bekk smoothness. However, it is pressure depression of the former room from which air is sent out that is measured with a water column manometer in Oken tester. The pressure depression is converted to time required to draw 10 ml of air at a pressure of 49.3 kPa, which is the definition of Bekk smoothness. Additionally, the length of air travel through the gap in the radial direction is 1 mm, while it is ca 17.8 mm for Bekk. This reduces a proportion of air permeating paper to the other side. In figures, the cube root of Bekk smoothness was plotted, because this dimension is equivalent to an inverse of Parker Print-Surf roughness[17].

Stress relaxation in compressive mode

In a compressive test, 7 to 9 sheets were stacked so that the total basis weight were ca. 480 g/m^2 (actually 444 to 590 g/m^2). A stack of sheets was inserted between the load cell and the cross bar head of a tensile tester UTM-Ⅲ-100 (Orientec co. ltd., Tokyo, Japan). To obtain even stress distribution, the compressed area was reduced by placing a coin-shaped piece of steel between the top sheet and the cross bar head. So, the compressed area was 707 mm^2. Figure 2 illustrates how to set the sheet stack. The compressing speed was constant at 0.667 mm/s.

RESULTS AND DISCUSSION

Load-compression curve of stacked paper sheets

Figure 3 shows an example of the load-compression curve. After a rapid increase in compressive load, the compressing was stopped and the load kept measured for 120 s. The slope of load decrease plotted against logarithmic time was linear after ca. 0.5 s. Logarithmic time law[4] approved in the tensile mode of paper can apply to the compressive mode. Slopes of the load decrease, namely, relaxation speed, for varied initial stresses were measured and the relation between them (Kubát's plot) consisted of two linear portions, also as well as found in the tensile mode.

Calendering effect on transverse internal stress

Figure 4 shows the relaxation speed versus the initial stress, what we call Kubát’s plot in the compressive mode. Every relationship traced a steep rectilinear slope at lower initial loads and then a gentle rectilinear slope at higher ones. The steep slope did not necessarily go through the origin, but intersected the relaxation speed axis at a positive value. Those characteristics resembles those of Kubát’s plot in the tensile mode, though the relations are all reversed as seen by comparison to Figure 1. Calendered sheets showed lower relaxation speeds than uncalendered ones. But, the calendering effect seemed to be equivalent between at 29 kN/m and at 49 kN/m. Water treatment, that is, soaking in water for 1 day and redrying, shifted the three relationship upward, namely, in the direction of increasing relaxation speeds, to an almost common relationship . According to Kubát’s theory for the tensile mode, the intercept of the later rectilinear slope in extension on the initial stress axis is interpreted as an in-plane internal stress. This is possible because every slope is in parallel and has a constant gradient of ca. 0.1, irrespective of in-plane internal stress level. However, the corresponding slope in the compression mode, namely, the gentle rectilinear slope showed a fairly different gradient for the water-treated sample from those unsoaked. For the present, we defined the initial load with the relaxation speed equal to 25 N/*log(s)* in the gentle slope ranging from 294 to 687 N (30 to 70 kgf) or in its extension as an index of the transverse internal stress. The transverse internal stress so defined are tabulated also in Table 2. The value of 25 N/*log(s)* does not have a special meaning but close to the mean value of the measured range. Even for the tensile mode, the slopes empirically determined are not always in parallel accurately, so a shift distance at certain relaxation speed can be more reasonable to estimate a change in in-plane internal stress. The gradient of the gentle slope was attempted to the analysis, but did not result in a systematic relation.

Wet-pressing effect on transverse internal stress

Figure 5 shows a Kubát's plots for the handsheets of the wet-pressing series. It is clearly shown that the stronger the wet-pressing, the lower the relaxation speed. Based on the transverse internal stress defined in the above section, it followed that wet-pressing increased the transverse internal stress. Water application decreased the transverse internal stress except for 49 kPa for 1 min. This suggests that the network and fiber structures formed at the lowest level of wet-pressing seems not to have been changed by soaking and drying. For the other two higher levels, the reductions in transverse internal stress were much less than those for the calendered wood-containing papers.

Beating effect on transverse internal stress

Figure 6 shows a Kubát's plot for the handsheets of the beating series. The higher the beating degree, the higher the transverse internal stress. It was interesting that the unbeaten sample shows one straight line throughout the full range of initial load. This implies that the transverse stress of uncollapsed fibers relaxed in an invariable manner throughout the range. Water application overall tended to reduce transverse internal stress. The unbeaten sample with the lowest transverse internal stress showed a fairly large reduction by water application, which is different from the case of the least wet-pressed sample. Generally, beating increases flexibility of fibers in water and contributes to formation of intra- and interfiber bondings. Wet-pressing assists, in particular, interfiber bondings. For the unbeaten sample intermediately wet-pressed, the fibers are considered to recover a little to tube-like shape by water application, lowering the relaxation speed. For the least wet-pressed sample intermediately beaten, in contrast, the fiber shape is considered to be hardly changed by water application [15], keeping the relaxation speed. However, the shape of the fibers should be confirmed in other study.

Filler loading effect on transverse internal stress

Figure 7 shows a Kubát's plot for the handsheets of the filler loading. The three relationships were all similar and slight differences seem to be within experimental errors. It was found that filler loading did not affect transverse internal stress. This finding suggests that fewer interfiber bondings due to a larger amount of filler loading are not related to the transverse internal stress. Collapse of fibers and resulting fiber stiffness, rather, determines the transverse internal stress. The unloaded sample was equivalent to intermediately beaten and intermediately wet-pressed ones shown in other figures, but the total basis weight of the tested stack was higher for the unloaded sample. This is the reason for a lower transverse internal stress for it. For the present, experiments of water application have not been made with this series.

Relationship between transverse internal stress and smoothness

Values of transverse internal stress for all the samples used are listed in Table 2. It is considered that, for the conditions used, beating had the largest effect on increasing transverse internal stress. Then, calendering, the second, and wet-pressing, the last though the values are not exactly corrected based on basis weight. Figure 8 shows the relationship between transverse internal stress and the cube root of the mean smoothness of the two sides. The lines connecting every two points shows changes caused by water application. The point on the lower-left side always denotes a value after water application. For all of the kraft handsheets except the filler loading series (left out because of the higher total basis weight), the higher the transverse internal stress, the higher the smoothness. The calendered wood-containing paper also shows the same tendency, but the smoothness was lower than those of the kraft handsheets at equivalent transverse internal stresses. Furthermore, the reduction of transverse internal stress was much larger than those of the kraft handsheets. In the case of mechanical pulp the shape recovery, ribbon-to-tube, of fibers are generally considered to be a main cause for surface roughening[14]. From this standpoint, this shape recovery by water application may have a strong influence on reduction in transverse internal stress.

Figure 9 shows index of transverse internal stress versus apparent sheet density for every sample used in this work. Past literature taught that a procedure of compression of a fiber network structure was well formulated with regard to density as shown[18] in Equation (1):

$$\rho^3 - \rho_0{}^3 = MP^N \qquad (1)$$

, where ρ, ρ_0 and P are the density, the density under no pressure and the pressure, respectively. M and N are constants. Historically, a similar equation applied to water saturated pulp[19] and a wool fiber mass[20]. Other modified formula was also tried to paper board[21]. For kraft handsheets and post-water application calendered wood-containing paper, transverse internal stress are related to apparent density as all the data points fall on the same curve. Only the data points for pre-water application calendered wood-containing paper do not meet this relation. Certain different mechanism should have worked with those samples.

Behavior of paper in compressing process

What was occurring during the compressing process and during the stress relaxation process? It was assumed as follows: Initially, small interfiber voids made by relaxation right after wet-pressing and calendering were being filled with adjacent fibers again[22]. At the same time, fibers were becoming bent and incompletely collapsed fibers were moreover collapsing. At higher loads, inter-lamella or inter-fibrillar voids in fiber walls were collapsing. During the relaxation process, in addition to fibers packing, fiber bending, fiber collapse and fiber wall collapse that all continued to occur, viscous deformation - viscose flow of the material comprising fibers toward less-stressed portions - was presumably proceeding.

Yanagida[23] divided the hot-pressing process in forming a particle board into several stages; packing of apart chip elements with mutual collisions and frictions; resistance of the packing; bending, buckling and compressive deformation of individual elements; and in-plane expansion of the board. The stages were clearly distinguished for an element-aligned structure, but unclear for a random structure. The author found the last stage above ca. 9.8 MPa, while the maximum of the stress in this work was up to ca. 0.97 MPa. So, only deformation of the packing level and element level (namely, the fiber level in the case of paper) should be involved in the compressive stress relaxation in this work.

Jackson and Ekström measured relative compression (total compression at maximum pressure to initial paper thickness ratio) and relative expansion (elastic recovery after pressure removal to the total compression) for various kinds of paper. Their results showed that beating reduced the relative compression, the effect of clay filling on the compressibility was negligible, coating suffered heavy reduction in the compressibility, and overdrying at 100 ºC reduced the compressibility. They considered that a stiffening of the cell wall caused a reduction in the relative compressibility but an increase in its elastic recovery. The compressive stress relaxation in this work harmonized well with their results as, for example, beating reduced the relaxation speed because it is a phenomenon resulting from both compression and expansion. They stated that there is some relationship between surface smoothness and compressibility characteristics, it is important to realize that this relationship will be different among papers from different pulping processes. Paper with high transverse internal stress are commonly composed of highly collapsed fibers by beating, wet-pressing and calendering. Such a sheet has little intrafiber void (lumen) to allow viscose flow for relaxation. Particularly, beating makes fibers stiff, resulting in less pliability. So, those transversely operated processes increased transverse internal stress. In comparison in relation to smoothness between fiber sources, mechanical fiber or chemical fiber, there was other limitations involved. One was machine-made and the other was hand-made. Besides, the comparison must be made for the same papermaking process. More exact interpretation needs further researches.

CONCLUSIONS

A new concept was proposed that what dominates surface roughening is not in-plane internal stress built in drying, but transverse internal stress built in transverse processes such as calendering and wet-pressing.

The analytical procedure of Kubát proposed for in-plane internal stress was attempted to apply to the transverse compressive mode to determine the transverse internal stress. A straight-line portion was found at higher initial stresses in the plots of relaxation speed per unit logarithmic time versus initial stress (Kubát's plot). The slope and the offset of this straight-line portion were varied with the process the sheet subjected. The initial stress at a relaxation speed of 25 N/*log(*s*)* was defined as an index of transverse internal stress for convenience, though theoretical consideration is further needed. Consequently, the greater the calendering, wet-pressing and beating, the higher the transverse internal stress (the lower the relaxation speed). But, the amount of loaded calcium carbonate did not affect it. The transverse internal stress had influence on surface roughening of paper, but the relationship was observed to be different between the papermaking processes. Besides, the transverse internal stress was likely to be associated with apparent sheet density depending on the fiber source and the way of sheet making. The authors considered that the degree of fiber collapse and viscoelasticity are closely related to the transverse internal stress.

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Process	Fiber source	Sheet-making	Composition	Condition
Calendering	Wood-containing	Machine-made	Softwood kraft/ GWD/TMP =15/40/45	Uncalendered 29 kN/m supercalendered 49 kN/m supercalendered
Wet-pressing	Wood-free	Handsheet	Hardwood kraft 100 %	49 kPa for 1 min 343 kPa for 5 min 686 kPa for 20 min
Beating	Wood-free	Handsheet	Hardwood kraft 100 %	Unbeaten, 639 ml CSF 5000 rev. PFI,460 ml CSF 20000 rev. PFI, 260 ml CSF
Filler loading	Wood-free	Handsheet	Hardwood kraft 100 %	Unloaded 10 % to dry pulp 30 % to dry pulp

Table 1 Furnish and process of paper samples used.

Process	Condition	Number of sheets	Total basis weight, g/m^2	Stack thickness, μm	Apparent density, g/cm^3	Transverse internal stress index, N
Calendering	Uncalendered	8	586	1087	0.540	649
	(soaked & dried)	8	591	1240	0.477	372
	29 kN/m	8	581	989	0.588	766
	(soaked & dried)	8	589	1184	0.497	364
	49 kN/m	8	588	941	0.624	743
	(soaked & dried)	8	597	1202	0.497	376
Wet-pressing	49 kPa, 1 min	8	493	946	0.521	401
	(soaked & dried)	8	487	970	0.502	397
	343 kPa, 5 min	8	447	752	0.595	555
	(soaked & dried)	8	444	808	0.550	505
	686 kPa, 20 min	8	445	689	0.647	698
	(soaked & dried)	8	448	749	0.598	623
Beating	Unbeaten	7	491	1059	0.463	345
	(soaked & dried)	9	465	1045	0.446	288
	5000 rev. PFI	7	453	765	0.592	521
	(soaked & dried)	8	444	808	0.550	505
	20000 rev. PFI	7	448	620	0.722	867
	(soaked & dried)	7	444	645	0.689	722
Filler loading	Unloaded	8	522	849	0.615	407
	10 % to dry pulp	8	549	894	0.614	399
	30 % to dry pulp	8	545	899	0.606	441

Table 2 Physical properties of samples used and transverse internal stress defined as an initial load at 25 (N/log(s)) of relaxation speed.

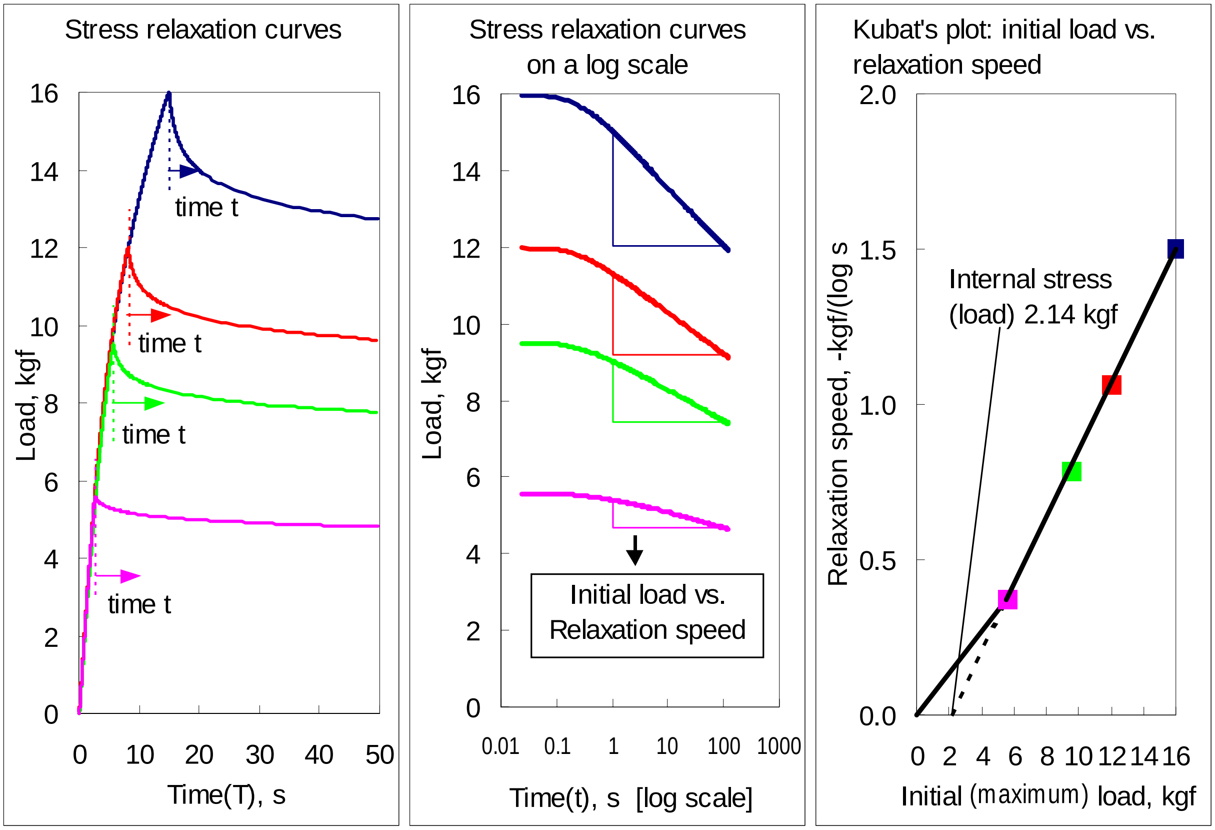

Fig.1 Procedure to determine internal stress in tensile mode by Kubat's method.

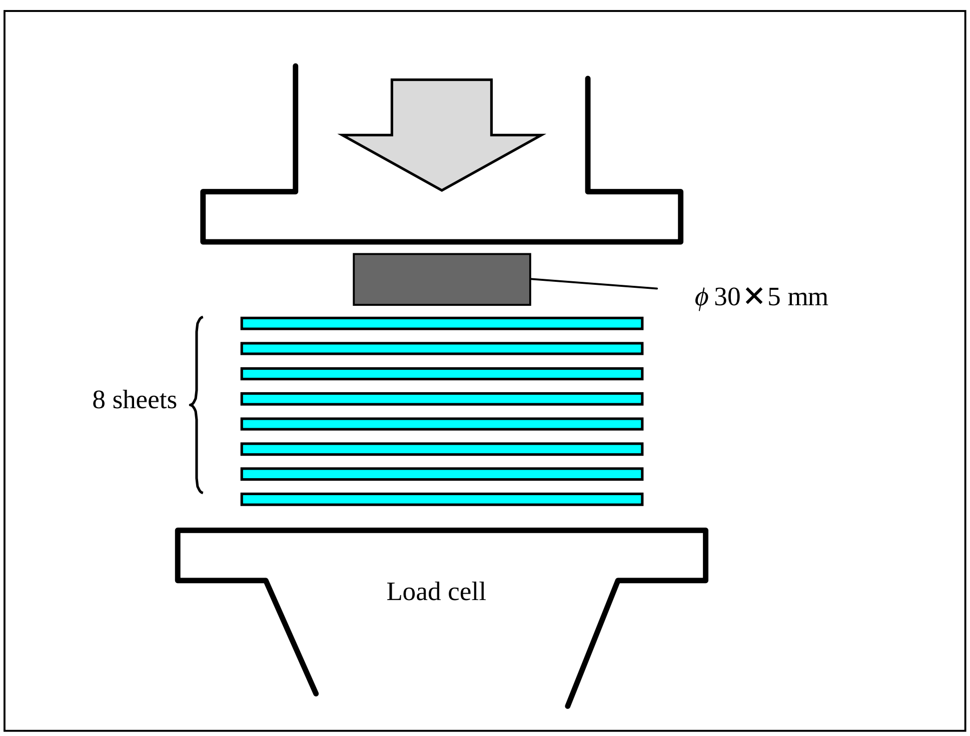

Fig. 2 Schematic diagram of sheet stacking in the compressive tester.

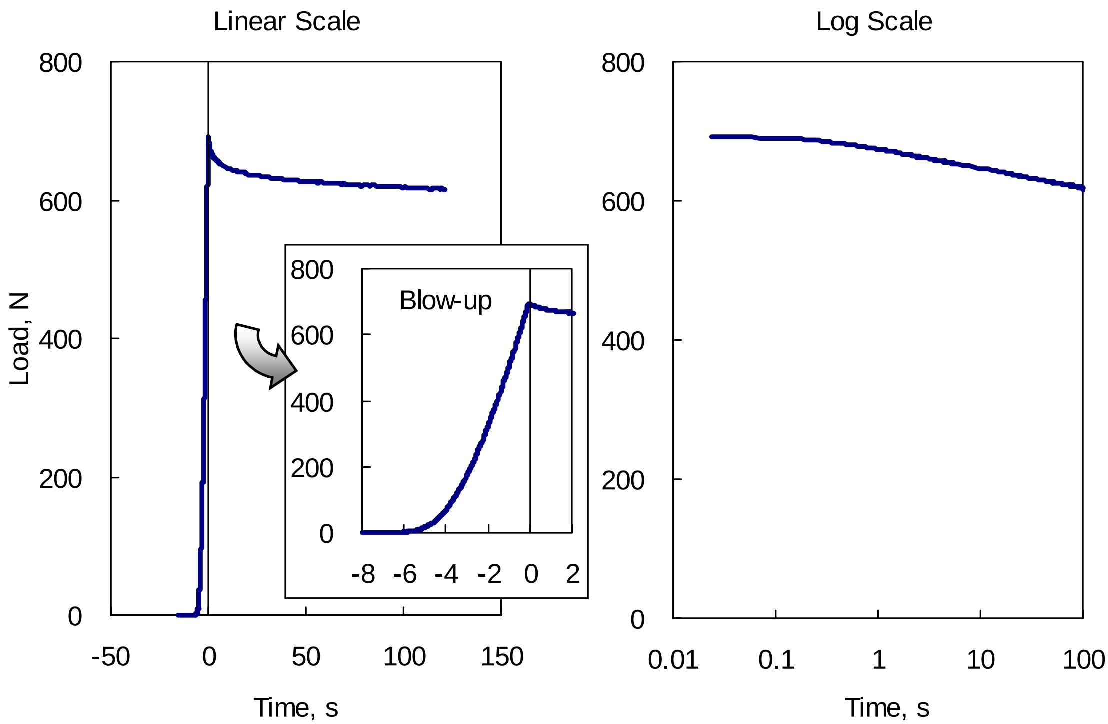

Fig. 3 Load-compression curve and procedure to determine relaxation speed of compressive stress.

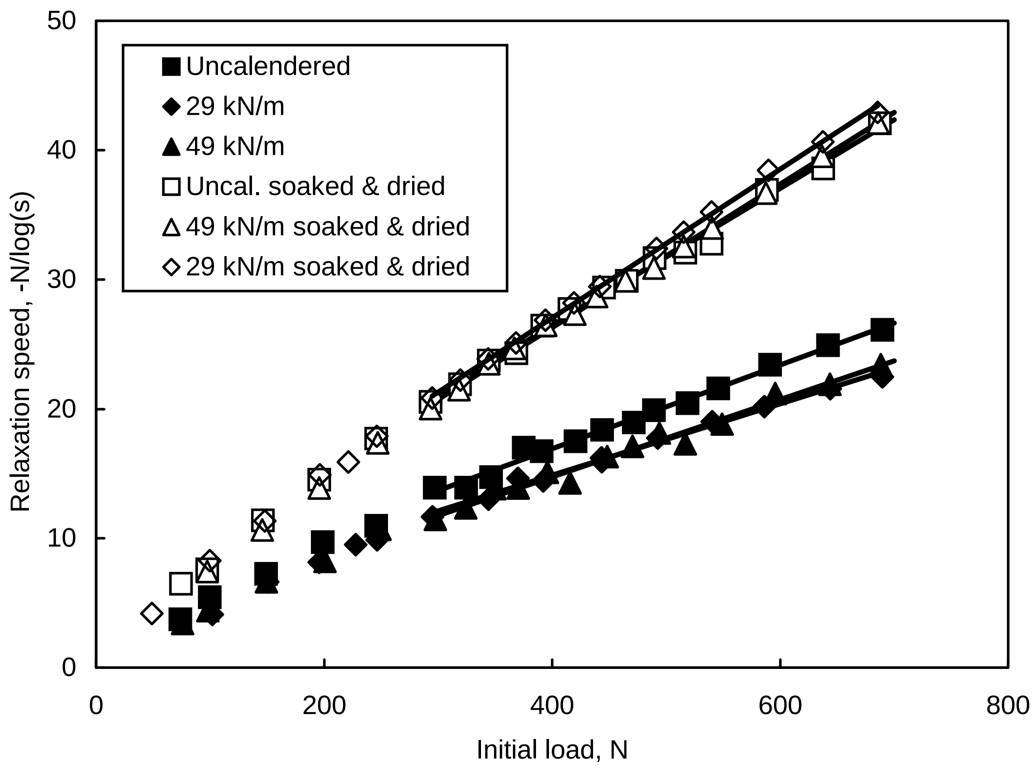

Fig. 4 Relaxation speed of compressive stress vs. initial load (Kubát's plot) for wood-containing sheets variously supercalendered.

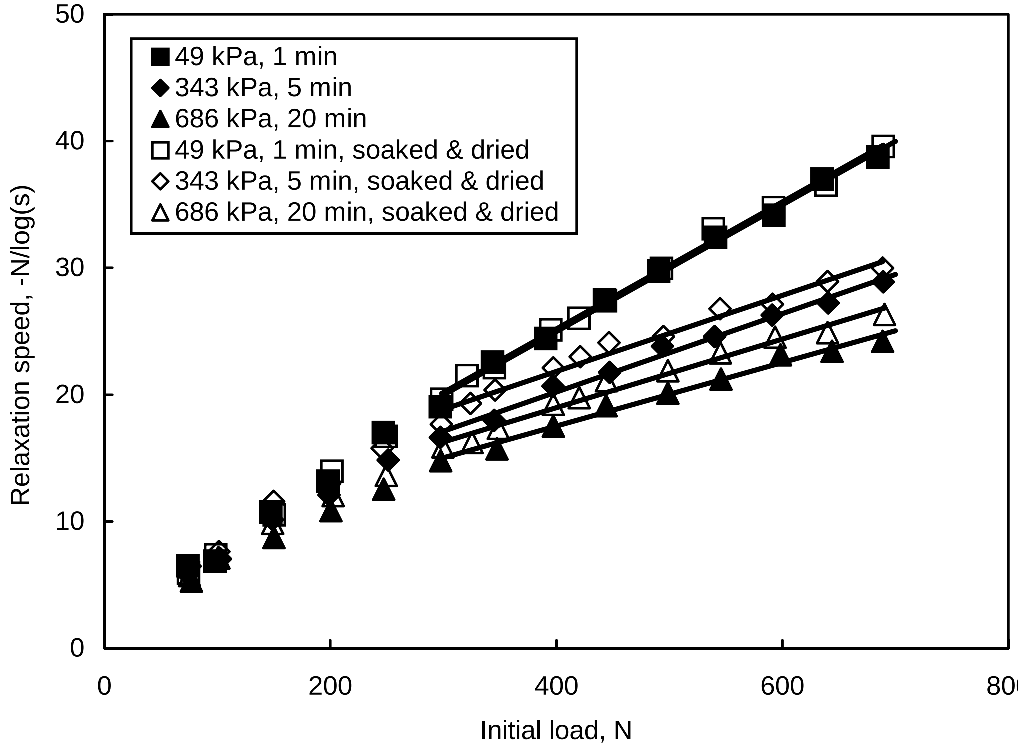

Fig 5. Relaxation speed of compressive stress vs. initial load (Kubát's plot) for handsheets variously wet-pressed.

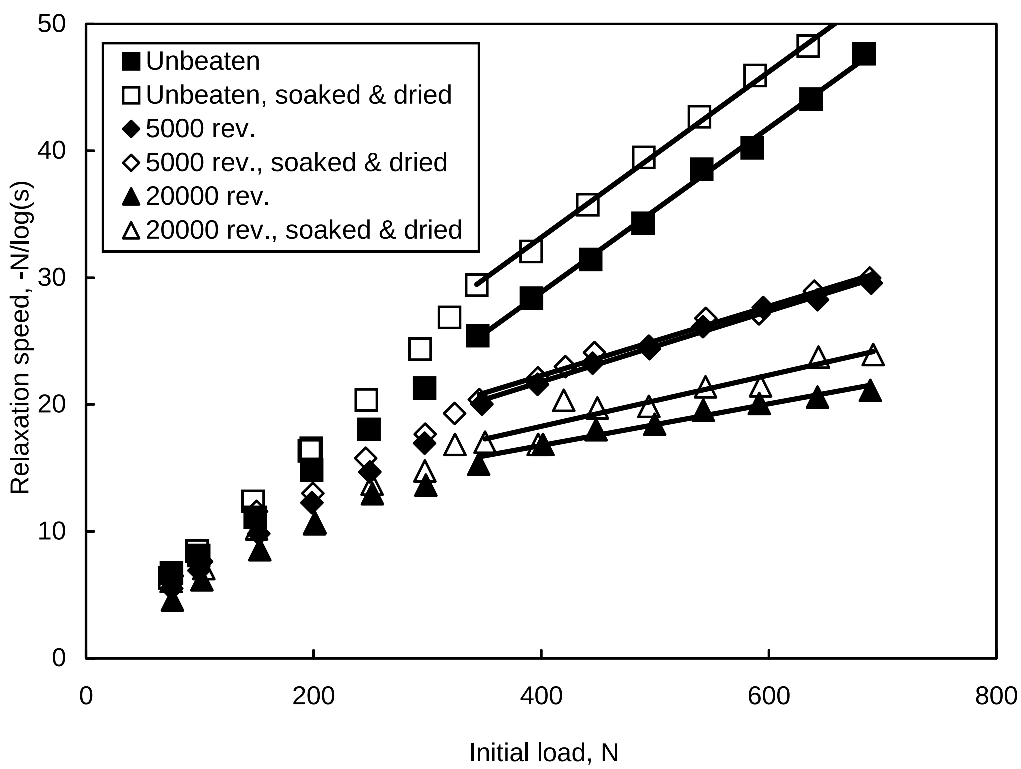

Fig. 6 Relaxation speed of compressive stress vs. initial load (Kubát's plot) for handsheets of different revolutions in beating.

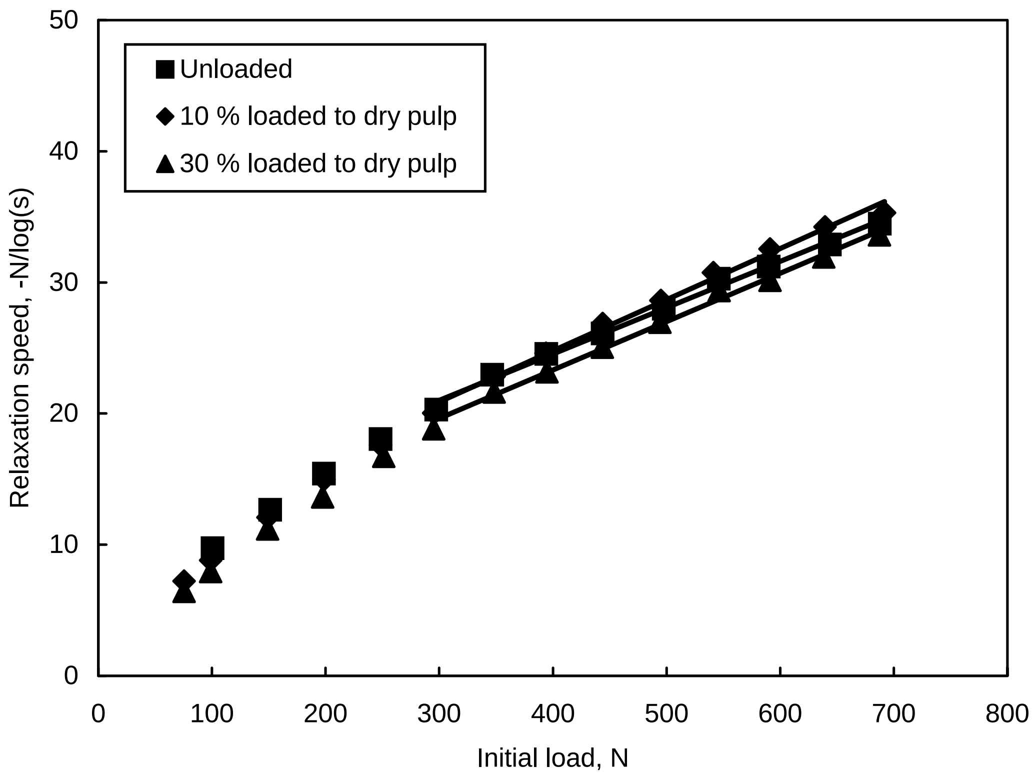

Fig. 7 Relaxation speed of compressive stress vs. initial load (Kubát's plot) for handsheets loaded with calcium carbonate.

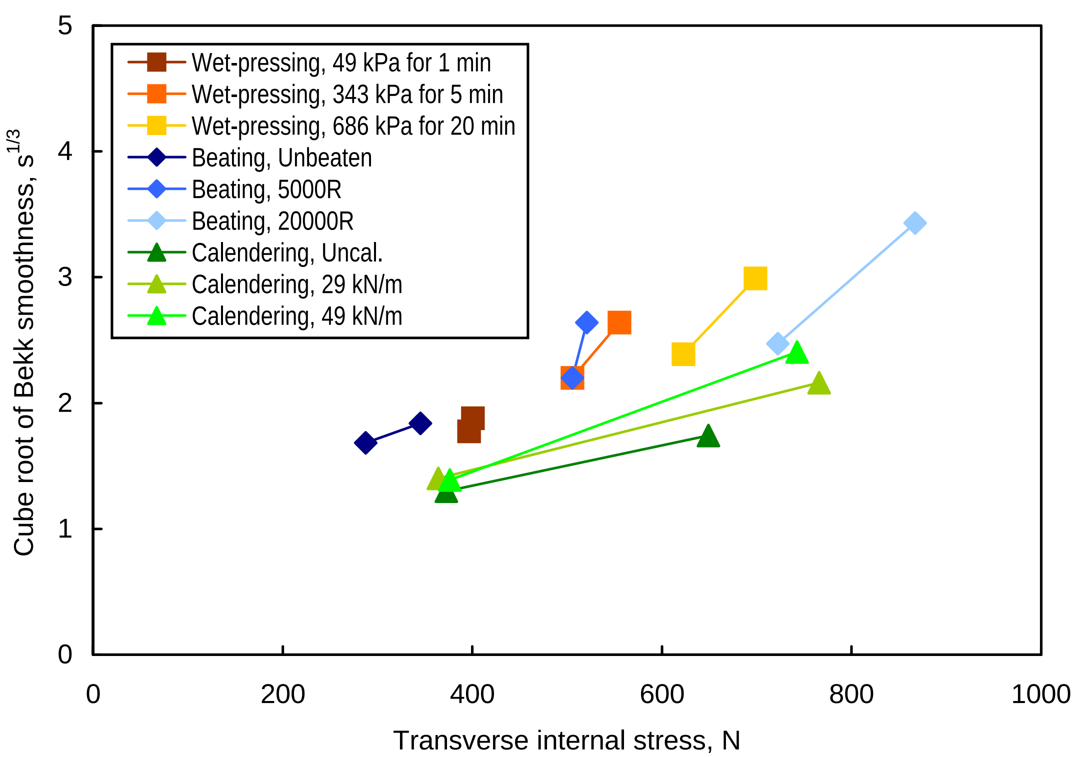

Fig.8 Relationship between transverse internal stress determined by Kubát's method and Bekk smoothness.

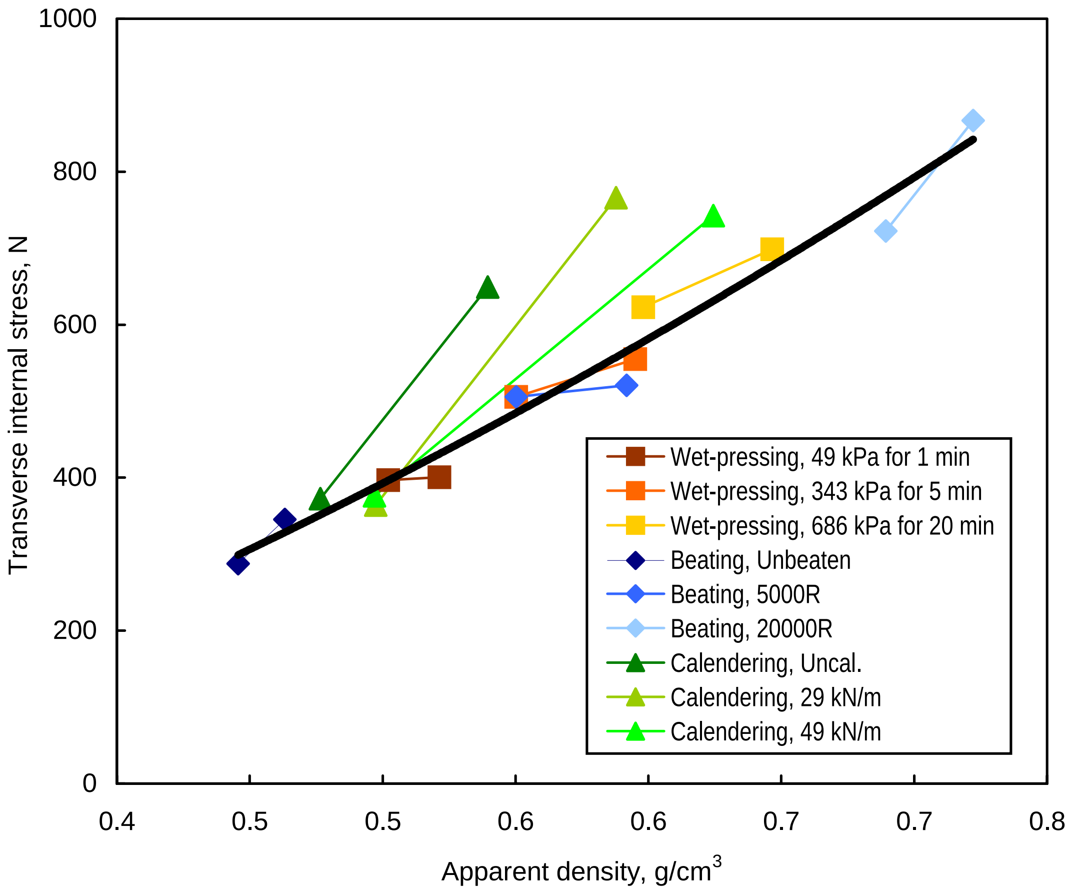

Fig.9 Relationship between transverse internal stress determined by Kubát's method and Apparent sheet density..