

**Original Paper** ~~~~~

## **Effects of Repeated Drying-and-rewetting and Disintegration Cycles on Fundamental Properties of Dissolving Pulp Fibers and Paper Made from Them**

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In order to study paper strength reduction on recycling and the effect of hemicellulose on the decrease of specific bond strength during recycling, handsheets made from prehydrolyzed kraft pulp for dissolving were repeatedly recycled up to 30 times with and without disintegration. The results were compared with those from kraft pulp for papermaking. The effects of recycling with and without disintegration on the fundamental properties of dissolving pulp fibers and paper made from them were quite similar to those of papermaking pulp. The effect of hemicellulose on the specific bond strength with recycling may be negligible. The decrease of external fibrillation due to disintegration during paper recycling affected CSF and WRV and could cause a decrease of specific bond strength, resulting in further strength reduction of paper after several times recycling.

**Keywords:** Paper recycling, Strength reduction, Disintegration, External fibrillation, Hornification, Dissolving pulp, Papermaking pulp

### **Introduction**

One of the most characteristic properties of paper material is its high ability to be recycled; the disintegration, molding, drying and rewetting of paper are easily repeated. Thus, almost all paper material for packaging use is made from recycled pulp. However, the strength of paper made from kraft pulp remarkably decreases through recycling because the recycling causes fiber hornification, loss of external fibrillation and so on<sup>1,2)</sup>. In the first report of a serial study on paper recycling<sup>3)</sup>, handsheets made from softwood and hardwood bleached kraft pulps for papermaking were repeatedly subjected to only drying-and-rewetting cycles up to 30 times and properties of pulp fiber and the paper made from them were compared with those recycled through the drying-and-rewetting and disintegration processes in order to study relation between the strength reduction and hornification, since the “net hornification” is caused only by repeated drying-and-rewetting without disintegration. The results suggested that decrease in external fibrillation with increasing recycle number might have a large effect on the changes in freeness (CSF) and water retention value (WRV) and further could partly explain the large decrease in tensile strength of the paper made from recycled pulp with disintegration. In the second report of the serial study<sup>4)</sup>, a further reduction of the strength at many times recycling with disintegration could be partly attributed to decrease in specific bond strength. In this study, taking into account the role of hemicellulose in

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fiber-fiber bonding and specific bond strength, handsheets from prehydrolyzed kraft pulp for dissolving, the hemicellulose content of which is low, were recycled in the same way as in the previous study<sup>3)</sup> and the fundamental properties of pulp fibers and papers made from them were examined and compared with those from papermaking pulp.

## Experimental

**Materials**<sup>3)</sup> As the base pulp a commercially available prehydrolyzed kraft pulp for dissolving use, in the form of dry pulp sheet, was used without classification to remove the fines fractions in this study. The pulp ( $\alpha$ -cellulose content:91%) was beaten with a PFI mill to 115 ml in CSF. Handsheets of 60 g/m<sup>2</sup> were first made from the pulp according to TAPPI Test Method T 205. After taking some sheets to provide a sample before recycling, the remainder were soaked in water overnight, and then re-dispersed for 25 min with a standard laboratory disintegrator. After measurement of CSF and WRV, handsheets were molded, wet-pressed and dried overnight with drying rings. The drying condition was an air blowing at 60°C to avoid heat deterioration. Recycling without disintegration was also conducted. These recycling procedures were repeated 30 times with sampling for some sheets at 1, 2, 3, 5, 10, and 30 cycles.

## Measurement

The water retention value (WRV) of wet handsheets was determined according to Japan TAPPI Test Method 26 (3000g, 15min). Other fiber properties except CSF were measured optically using a fiber quality analyzer (OpTest Equipment Inc.). All dry handsheets before and after recycling were thoroughly conditioned at 23°C and 50% relative humidity, and the following physical measurements were made. The thickness value needed to determine density and Youngs modulus was measured by the rubber platen method. The tensile strength and Youngs modulus were measured using an InstronType machine (Shimadzu Autograph GS-100) with a pair of line type PAPRICAN clamps<sup>5)</sup> to secure a measurement under in-plane stress loading. Tear strength was measured using the same tensile tester<sup>6)</sup>. The measurement of zero-span tensile strength as a measure of fiber strength was performed using the IPC

Table 1 Fundamental properties of the pulp fibers before and after thirty times recycling with and without disintegration

	Percent fines <sup>*1</sup> %	Coarseness mg/100m	Mean fiber length <sup>h*2</sup> weight-weighted	Mean curl length-weighted	Mean kink kinks per mm	Zero-span strength Nm/g
Before recycling	17	16.0	1.2mm	0.07	0.37	68
After recycling						
With disintegration	4	15.7	1.2mm	0.06	0.38	70
Without disintegration	17	16.7	1.2mm	0.07	0.38	64

<sup>\*1</sup>:less than 200  $\mu$  m, <sup>\*2</sup>:70  $\mu$  m—10mm

type attachment. Optical reflectance of the paper with black velvet backing ( $R_0$ ), and that with a backing consisting of a pad of the same papers ( $R_\infty$ ) were measured at several points on the specimen using a photovolt type reflectometer (Tokyo Denshoku TC-6D). The scattering coefficient was calculated from the values of the measured  $R_0$ ,  $R_\infty$ , and basis weight using the Kubelka-Munk equation.

## Results and discussion

### Change in fundamental properties of pulp fibers before and after recycling

The fundamental properties of the pulp fibers before and after 30times recycling are shown in Table 1. On recycling almost no change in the properties including fiber length and fiber strength (indexed as zero-span strength) was found, regardless of whether disintegration was included or not. The exception was a marked reduction of fines fraction by recycling with disintegration, which is natural as a result of not having carried out screening with a classifier in advance.

Freeness (CSF) unexceptionally increased with increasing recycle number, although the rate of increase and its detail differed with variations in sample and recycling conditions. The increase of CSF in this experiment was sharp in the early recycling stages and gradually decreased with increasing recycle number as shown in Fig.1. The change in internal (fiber wall) fibrillation by recycling is commonly referred to as hornification and WRV is used as a fiber hornification index. The decrease in WRV with increasing recycle number is shown also in Fig.1 as a percentage decrease ( $100 \times (\text{WRV before recycling} - \text{WRV at nth recycling}) / \text{WRV before recycling}$ ) with and without disintegration. WRV decreased rapidly at the early recycling stage and gradually leveled off with further increase in recycle number. The WRV decrease of pulp without disintegration was larger than that of pulp with disintegration at the early recycling. Since the change in WRV by recycling without disintegration is thought to be a net change of hornification during the drying-rewetting cycle, the disintegration might have an additional reverse effect on the decrease of WRV. One explanation of the reverse effect could be that a loss of external fibrillation by disintegration contributes to the difference of WRV between with and without disintegration, as suggested in the previous study for papermaking pulp<sup>3)</sup>.

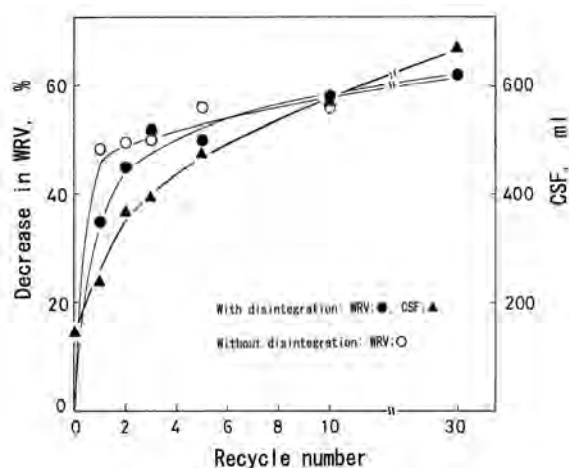


Fig.1 CSF and decrease in WRV as the percent decrease with increasing recycle number.

### Effect of recycling on the fundamental properties of the papers

The percentage changes in the density and other properties, defined as  $100 \times (\text{property value before recycling} - \text{value at } n\text{th recycling}) / \text{value before recycling}$ , are shown against recycle number in Fig.2 (with disintegration) and Fig.3 (without disintegration). As to the papers from recycled pulp with disintegration, decreasing behaviors of density, tensile index and Youngs modulus with recycle number were similar each other, i.e., the decrease of these on the first and second recycling was eminent. In contrast, the decrease of density with recycling was quite small for papers from recycled pulp without disintegration, while the hornification or reduction of wet fiber flexibility due to repeated drying-and-rewetting should be the same as that with disintegration, as suggested from the small difference in WRV between with and without disintegration. Further, the decrease of tensile index and Youngs modulus was smaller than that for paper from recycled pulp with disintegration. The difference between with and without disintegration was eminent in early recycling.

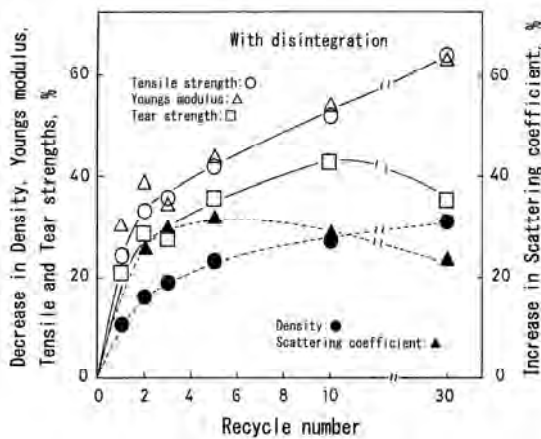


Fig.2 Decrease in density, Youngs modulus, Tensile and Tear indexes and increase in scattering coefficient as percent changes with increasing recycle number for the papers made from recycled pulp with disintegration.

### Relationship between Youngs modulus and tensile strength

Fiber bonding is a key factor in paper strength. Thus, tensile strength correlates very well with Youngs modulus. Further, Youngs modulus, which represents mechanical fiber bond strength, could have a linear relation with the mechanical fiber bonding area, assuming that the specific bond strength (bond strength per bonded area) is independent of beating degree and constant. A slightly curved linear relation was found for papers from papermaking pulp, regardless of disintegration<sup>4)</sup>. The relationship between Youngs modulus and the tensile index of the papers from recycled dissolving pulp generally gave a similar slightly curved linear relationship, as shown in Fig.4. However, the strength of papers from recycled pulp without disintegration, excluding paper recycled 30 times, was lower than that with disintegration, as long as the Youngs modulus level was the same. The result suggested that a factor other than mechanical fiber bonding, e.g. fracture toughness, was additionally related to tensile strength. Tear strength, as a substitute for fracture toughness, of the papers tested is discussed below.

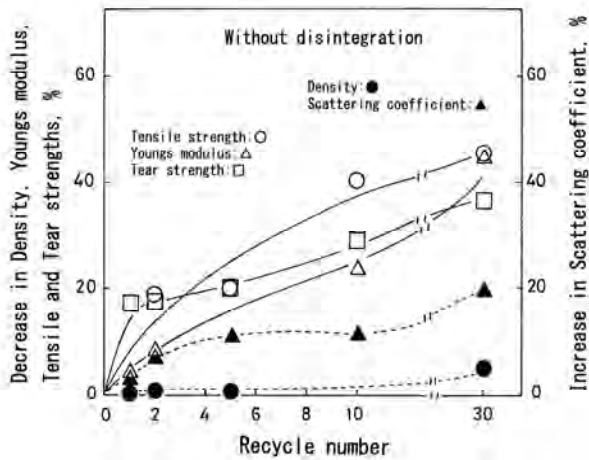


Fig.3 Decrease in density, Youngs modulus, Tensile and Tear indexes and increase in scattering coefficient as percent changes with increasing recycle number for the papers made from recycled pulp without disintegration.

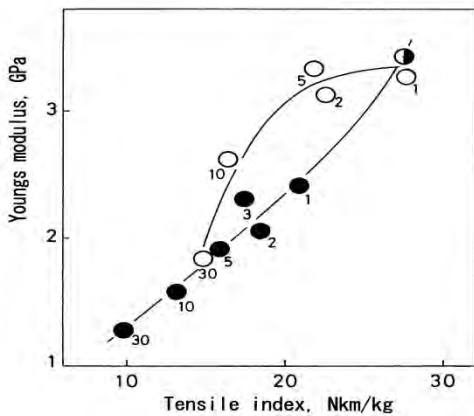


Fig.4 Relationship between Youngs modulus and tensile index for papers made from recycled pulp for dissolving with (solid symbol) and without (open symbol) disintegration: Figures indicate recycle number.

### Relationship between tear strength and tensile strength

The relationship between tear index and density of the papers from recycled dissolving pulp is shown in Fig.5. Incidentally, the tear index of papers from virgin and recycled LBKP for papermaking increases with density or tensile strength and levels off at ca. 9 Nm<sup>2</sup>/kg<sup>6</sup>). Although the tear index of paper from dissolving pulp was generally smaller than that of paper from papermaking pulp, the tear index gradually increased with increasing density for paper from recycled pulp with disintegration and rapidly increased with increasing density for papers made from recycled pulp without disintegration. Thus, the tear index of paper made from recycled pulp without disintegration is lower than that with disintegration, at the same

level of density. Taking into consideration of no difference in fiber length between with and without disintegration, the lower tear index or fracture toughness could be a possible additional cause of strength reduction for paper made from recycled dissolving pulp without disintegration.

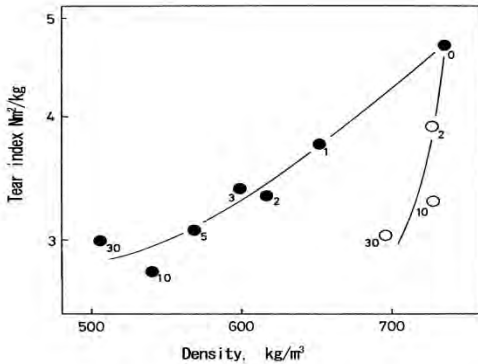


Fig.5 Relationship between tear index and density for papers made from recycled dissolving pulp with (solid symbol) and without (open symbol) disintegration: Figures indicate recycle number.

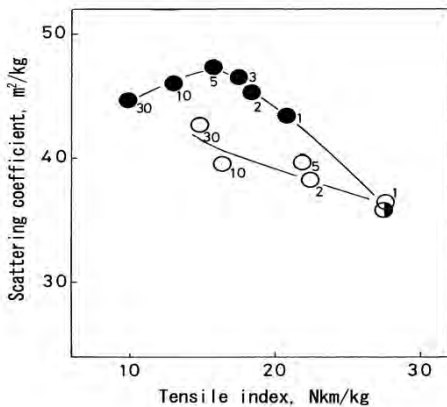


Fig.6 Relationship between scattering coefficient and tensile index for papers made from recycled dissolving pulp with (solid symbol) and without (open symbol) disintegration: Figures indicate recycle number.

### **Relationship between scattering coefficient and tensile strength**

The mechanical fiber bonding area is surrounded by the optical fiber bonding area (optical contact). Further, non-bonded area spreads outside of the optical fiber bonding area. The non-bonded area can be optically measured and is expressed as the scattering coefficient of the Kubelka- Munk equation. Thus, a decrease of the coefficient means an increase in optical fiber bonding area and further an increase in mechanical fiber bonding area. In fact, an increase in tensile strength accompanied a decrease in the coefficient for a series of papers made from papermaking pulp beaten to various degrees and papers made from recycled pulp without disintegration <sup>4)</sup>. Fig.6 shows the relationship between tensile index and the

coefficient for papers made from recycled dissolving pulp. As to the papers made from recycled pulp without disintegration, a decrease of tensile index caused by recycling accompanied an increase in scattering coefficient, regardless of recycle number. In contrast, the scattering coefficient of papers made from recycled pulp with disintegration increased with a decrease of tensile index caused by recycling up to 5 times and then decreased with further recycling from 5 to 30 cycles. The same behavior occurred for papers made from recycled papermaking pulp with and without disintegration and thus, the strength reduction at many times recycling could be partly explained by a decrease in specific bond strength<sup>4)</sup>. The similarity between papers with and without disintegration from both pulps for papermaking and dissolving suggested that the effect of hemicellulose on the decrease of specific bond strength was quite small. That is to say, cause of the decrease in specific bond strength due to multiple recycling could be mainly attributed to the decrease of external fibrillation caused by disintegration during recycling<sup>3,4)</sup>.

## **Conclusions**

The effects of recycling with and without disintegration on the fundamental properties of dissolving pulp fibers and paper made from them are similar to those of papermaking pulp. The decrease of external fibrillation due to disintegration during paper recycling caused a decrease of specific bond strength, which resulted in further strength reduction of paper after several times recycling.

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## **References**

- 1) Y. Yamagishi and R.Oye, *Jpn Tappi J.* 35(9) 787(1981)
- 2) M.Garg and S.P. Singh, *Appita J.*59(4)274(2006)
- 3) T. Yamauchi and M. Yamamoto, *Appita J.* 61(5)396(2008)
- 4) T. Yamauchi and M. Yamamoto, accepted for publication in *Appita J.*
- 5) R.S.Seth, A.G.Robertson, Y.W.Mai, and J.D.Hoffman, *Tappi J.*76(2)109(1993)
- 6) T. Yamauchi and A.Tanaka, *J. Wood Sci.*48(6)532(2002)

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