

**Original Paper** ~~~~~

# **Strength Properties of Wet-combination Paper Containing Dry Strength Resin\***

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\*This paper was presented in part at the 21<sup>st</sup> Annual Meeting of the Packaging Science and Technology, Japan.

The effect of wet-combination on some basic strength properties was examined by using a series of wet-combined papers made from paper plies containing cationic polyacrylamide (C-PAM) dry strength resin. As the basic in-plane and out-of plane strength properties, tensile, compressive, and internal bond strength were examined. Microfailures, which are detectable as acoustic emissions (AEs) and occurred during the tensile deforming process, were further analyzed to investigate the effect of the paper ply combination on in-plane tensile strength. The results showed that wet-combination with paper plies containing C-PAM brought about an increase in the plastic deformation region with a gradual increase in microfailures and delayed the acute occurrence of microfailures just before the maximum load. The higher strain at breakage rather than the increase in fiber bonding led to an increase in tensile strength. Heterogeneous location of strong paper ply containing C-PAM, especially on one side, decreased the increment of the tensile strength. The compressive strength in the X-Y direction increased with increasing total C-PAM content, regardless of wet-combination, although the increment was small for papers in which the strong paper ply containing C-PAM was located at the center or on one side. Tensile strength in the Z-direction measured as internal bond strength was governed by that of the weakest paper ply, and wet-combination brought about a further decrease in the Z-directional tensile strength.

Keywords : Acoustic emission, Compressive strength, Dry strength resin, Internal bond strength, Microfailures, Strain at breakage, Tensile strength, Wet combination

## **1. Introduction**

In the manufacturing of paperboard, wet-combination has been exclusively employed for a long time, and application of a dry strength agent has also become popular to make up for the strength decrease due to an increased use of recycled fibers. However, the effect of wet-combination on the strength properties of paper has not been thoroughly examined. In the previous report <sup>1)</sup>, the effects of wet-combination of paper plies from lightly and heavily beaten pulps on some basic strength properties of paper was examined, and microfailure occurrence during the tensile strength testing process, detectable as acoustic emissions (AEs), were investigated. They found that the layered paper structure made by wet-combination brought about an increase in the period of the plastic deformation region, leading to a

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higher strain at breakage with an increase in tensile strength. Compared with homogenous paper made by pulp mixing, the presence of a strong paper ply in wet-combination, irrespective of its Z-directional position, may increase the period of the plastic deformation region with gradual increase in microfailure and thus delay the acute occurrence of microfailures just before the maximum load. Further, including strong paper plies on both outer sides could prevent the development of microfailures in paper sheet failure, leading to an increase in strain at breakage and ultimately to higher tensile strength. In contrast to tensile strength, compressive strength in the X-Y direction was simply determined by the content of highly beaten pulp, regardless of the manner of combination, i.e. wet-combination or pulp mixing. Tensile strength in the Z-direction measured as internal bond strength was governed by the strength of the weakest paper ply, and wet-combination brought about a further decrease in the Z-directional tensile strength. As a serial study on the effect of wet-combination with consideration of effective application of a dry strength agent, wet-combination was conducted with paper plies containing dry strength resin, slightly containing it and not including it as strong, less strong and weak paper layers in this study. Firstly, the basic strength properties of the wet-combination papers varying in total content of dry strength resin and in location of strong or less strong paper plies were examined. Secondly, microfailures occurred during the tensile testing process were investigated with a help of AE analysis as a fundamental study into the effect of wet-combination on the tensile strength properties.

## **2.Exerimental**

### **2.1Materials**

The cationic polyacrylamide (C-PAM) used in this study as a dry strength resin was a commercially available standard C-PAM (relative molecular mass:  $2 \times 10^6$ , charge density: 0.6 meq/g)<sup>2)</sup> from Arakawa Chemical Industries, Ltd. (Osaka, Japan) at 7% concentration solution. No further purification of C-PAM was conducted.

The pulp used in this study was a commercially available fully bleached hardwood (mixed species) kraft pulp and was in advance screened using a single compartment classifier with a 100-mesh screen<sup>3)</sup>. The fines-free pulp was lightly beaten with a PFI mill to 510 ml CSF.

Aliquots of the lightly beaten pulp were diluted with tap water to 0.3% concentration (at pH 7) and then an aqueous solution of the C-PAM, which was adjusted to 1.0% concentration with deionised water, was added to the furnish to produce the papers containing C-PAM in the range of its concentrations 0 to 2% (mass of C-PAM on o.d. paper). No other additives (such as alum) were added to the furnish to simplify the experiment. After the addition of C-PAM, the furnish was continuously stirred at room temperature for 30 min before sheet molding using the internal application method<sup>2)</sup> to mold handsheets as follows. Couched wet handsheets of 30 or 60 g/m<sup>2</sup> basis weight were made from the above mentioned furnish, according to TAPPI Test Method T-205. In the case of 30 g/m<sup>2</sup> sheet, the water level in the molding machine was kept at half the normal level to keep the fiber consistency on molding constant, and thus the sheet formation was nearly constant between handsheets of 30 or 60 g/m<sup>2</sup> basis weight. The couched wet handsheet was once or twice overlaid on the next wet handsheet just after drainage to

prepare the sample combination papers (final basis weight: ca 120 g/m<sup>2</sup>). Thus, various combinations of paper plies were prepared. In more detail, the first series of papers were made of the same furnish but varying in number of paper plies (number of combination), and the second series of papers were made of two furnishes with and without C-PAM, keeping their total C-PAM content constant and varying the location of the paper ply with C-PAM. A third series of papers (basis weight: ca 120 g/m<sup>2</sup>) varying in C-PAM content were directly made without the combination operation. All wet papers were dried with drying rings under restraint by air blowing at room temperature. The total C-PAM content retained in the papers was determined by nitrogen analysis (Total Nitrogen Analyzer TN-110, Mitsubishi Chemical Analytec Ltd.)<sup>4)</sup>.

## 2.2 Basic and strength properties of handsheets

The thickness for determination of sheet density and Young’s modulus was measured by the rubber platen method according to TAPPI test method T-551. For determination of the Young’s modulus and tensile strength, test specimens were strained using an Instron type machine (Shimadzu Autograph AGS-100) with a pair of line type PAPRICAN clamps<sup>5)</sup> to secure a measurement under in-plane stress loading. Measurements of edgewise in-plane compressive strength (short span compressive strength) and internal bond strength were conducted according to TAPPI test method T-826 and -541, respectively. Internal bond strength is the physical breaking energy with constant strain at breakage, and thus could be proportional to Z-directional tensile strength. As a result it is conventionally considered to be a substitute for Z-directional tensile strength<sup>6)</sup>. The basic and strength properties of the paper samples tested are given in Table 1.

Table 1 Basic properties of the tested papers

| Method      | Number of paper plies | Content of C-PAM % | Basis weight g/m <sup>2</sup> | Thickness μm | Density kg/m <sup>3</sup> |
|-------------|-----------------------|--------------------|-------------------------------|--------------|---------------------------|
| Combination | 3                     | —                  | 117                           | 181          | 647                       |
|             |                       | 0.4                | 119                           | 181          | 656                       |
|             |                       | 0.8                | 121                           | 184          | 655                       |
|             |                       | 1.7                | 121                           | 185          | 651                       |
|             |                       | 0.2                | 119                           | 191          | 626                       |
|             |                       | 0.5                | 118                           | 190          | 618                       |
|             |                       | 0.4                | 119                           | 187          | 636                       |
|             |                       | 1.0                | 119                           | 185          | 643                       |
|             |                       | 0.5                | 120                           | 188          | 642                       |
| Mixing      | 1                     | —                  | 122                           | 180          | 682                       |
|             |                       | 0.5                | 122                           | 172          | 711                       |
|             |                       | 0.9                | 123                           | 172          | 717                       |
|             |                       | 1.5                | 124                           | 173          | 717                       |

|   |                                      |
|---|--------------------------------------|
|  | : Paper ply not containing C-PAM     |
|  | : Paper ply containing C-PAM of 0.5% |
|  | : Paper ply containing C-PAM of 1.0% |
|  | : Paper ply containing C-PAM of 2.0% |

## **2.3 AE measurement**

The experimental arrangement for AE measurement during tensile straining and for analytical processing of the detected AE was essentially the same as in previous papers <sup>1,4</sup>). A commercially available piezoelectric AE sensor (Model AE 901U; NF Corporation, Yokohama, Japan) with a fundamental resonant frequency of 150 kHz was attached to the lower part of the specimen. The sensor and the specimen were pinched with a lightweight clip-type clamp across the thickness of the specimen. The sensor surface, as a result, contacted well with the surface of the specimen. The detected AE signal was then processed in an AE data processor (Model AE 9640; NF Corporation, Yokohama, Japan), and several parameters of each AE event including the time when the AE signal first crossed the threshold level set at 50 dB, the maximum amplitude (expressed in dB), and the tensile load were successively recorded on magnetic tape. The recorded AE data were further processed with a computer (AE analysis software 0963; NF Corporation, Yokohama, Japan) after the test. The relationships between cumulative AE event counts and tensile load were examined as the selected AE parameters in this study. A slight variation in tensile and cumulative AE event count behaviour among samples was found due to the fundamental characteristics of micro- and macro-failures, but the general trend of these results was clear <sup>7</sup>). AE study of the compressive strength and internal bond strength was not applicable, since setting of the AE sensor on the paper sample was difficult for both tests and both measurements were conducted at high-speed and thus the periods of their deforming processes were too short to examine the AE behavior.

## **3. Results and discussion**

### **3.1 Effects of total PAM content, wet-combination and position of strong paper ply containing C-PAM on basic and strength properties of papers**

As reported in a previous paper <sup>3</sup>), the effect of C-PAM addition on sheet density was small. In contrast, wet-combination brought about an evident decrease in the density for papers having homogenous Z-directional distribution of C-PAM regardless of the C-PAM content, although the total PAM content differed to some extent between one and three ply papers. This was probably because an incomplete overlaying accompanied with wet-combination brought about a slight increase in thickness and thus a decrease in density, as described in the previous report <sup>1</sup>).

The effects of wet-combination and location of the C-PAM contained paper ply on tensile strength, edgewise compressive strength and internal bond strength are shown in Fig.1a, b, c, respectively in relation to total C-PAM content. The two former show strength in the X-Y direction and the latter shows strength in the Z-direction of paper sheets. Furthermore, tensile strength and internal bond strength are strength in tension mode, while edgewise compressive strength is strength in compression mode.

As previously reported <sup>2</sup>), the increase in tensile strength with increasing C-PAM content of the paper was generally large for C-PAM content lower than 1% and quite small for higher content. That is to say, an increase in total C-PAM content does not yield a proportional increase in tensile strength, in contrast to the proportional increase in tensile strength with content of highly beaten pulp in the previous paper <sup>1</sup>). Homogeneous paper made by wet-combination of three plies had a higher tensile strength regardless of

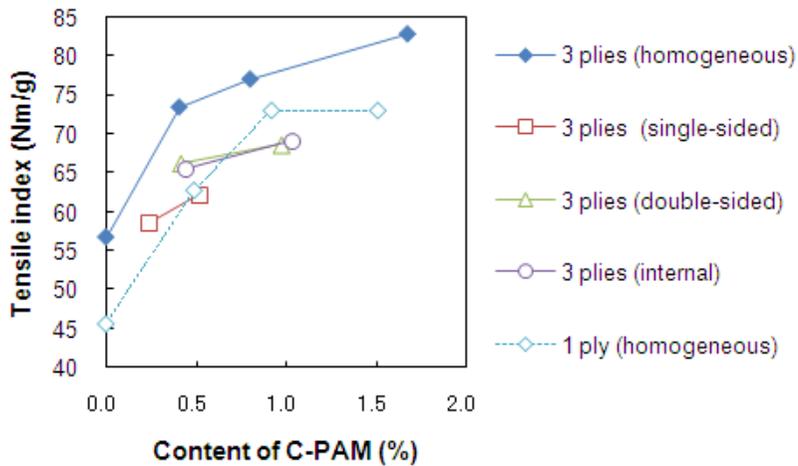


Fig. 1a Relationship between tensile index and total C-PAM content in the papers.

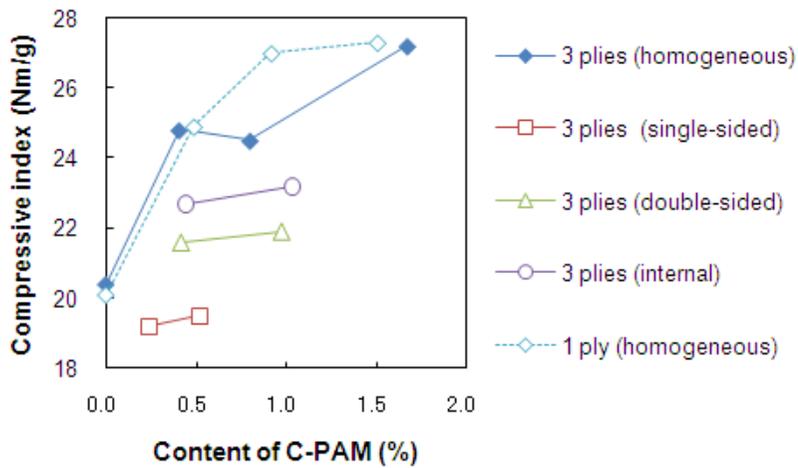


Fig.1b Relationship between compressive index and total C-PAM content in the papers.

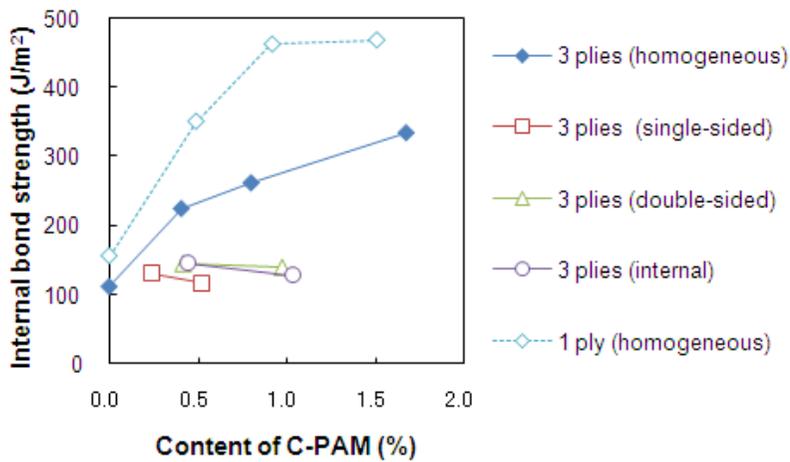


Fig.1c Relationship between internal bond strength and total C-PAM content in the papers.

the total C-PAM content, and the increment of tensile strength for lower C-PAM content was comparatively bigger than that for higher C-PAM content. A heterogeneous C-PAM location in the Z-direction (located at both outer sides or at one side) gave a lower tensile strength for the same total C-PAM content, suggesting a poor cost-performance of C-PAM addition.

Tensile strength had linear relationships with Young's modulus and strain at breakage as shown in Fig.2 a, b, respectively for all tested papers. Young's modulus, showing little variation between measurements, is a basic mechanical property and is essentially a physical constant of a solid material. Its increase means an increase of fiber-fiber bonding, leading to an increase in tensile strength. In contrast, strain at breakage is not a constant and shows some variation arising from the character of large deformation. Both generally showed linear relationships; the correlation coefficient for Young's modulus was 0.780 and that for strain at breakage was 0.913, and thus the latter was more decisive factor on the tensile strength, as mentioned in the previous paper <sup>1)</sup>. That is to say, the contribution of the elongation on tensile strength increase was bigger than that based on an increase in fiber bonding. The increase in tensile strength with increasing strain at breakage by wet-combination is further examined in the AE study in 3.2.

Edgewise compressive strength increased by C-PAM addition, and the increment was large for C-PAM content lower than ca 0.5 % and small for higher C-PAM content, regardless of wet-combination. Heterogeneous Z-directional C-PAM content by wet-combination gave a lower compressive strength than homogeneous Z-directional C-PAM content by wet combination for the same total C-PAM content. Notably in the case of one side location of strong paper ply containing C-PAM, the compressive strength was lower than that of paper containing no C-PAM. This result differed from the previous study <sup>1)</sup> in which compressive strength increased proportionally with the ratio of strong paper ply (from heavily beaten pulp) irrespective of the location of the strong paper ply.

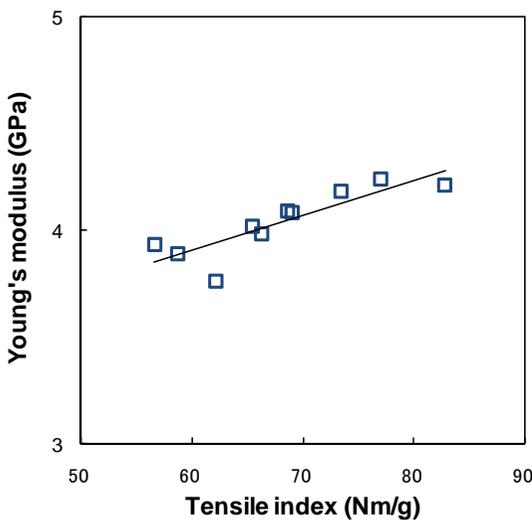


Fig. 2a Relationship between tensile index and Young's modulus for papers containing C-PAM.

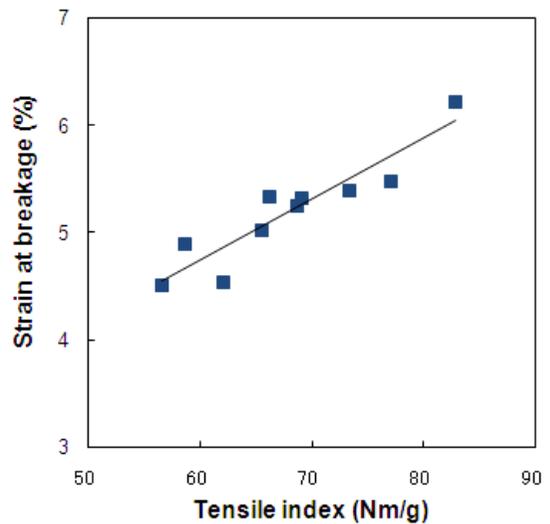


Fig. 2b Relationship between tensile index and strain at breakage for papers containing C-PAM.

The internal bond strength of papers not made by wet-combination steeply increased with the C-PAM content for C-PAM content lower than 1 % and leveled off at higher C-PAM content, while that of papers made of paper plies having the same C-PAM content by wet combination moderately increased with total C-PAM content. At any total C-PAM content, the former was higher than the latter. In contrast, the internal bond strength of papers made of paper plies having different C-PAM content by wet-combination was equal to or lower than that of paper containing no C-PAM. Thus, tensile strength in the Z-direction was determined by that of the weakest paper ply. Furthermore, incomplete overlaying accompanied by wet-combination could bring about some mechanical defects within the combined interface, easily leading to Z-directional sheet failure, i.e., low internal bond strength.

### 3.2 AE examination of papers made by wet combination (Effect of number of paper plies)

The occurrence of microfailures detected as AE events was examined from the relationships between tensile load and the cumulative AE event count up to the maximum tensile load, in order to investigate the effect of the number of paper plies in wet-combination. A representative result of papers made by wet-combination is shown in Fig.3 for papers made of paper plies containing C-PAM at total C-PAM content of ca. 1 %. Little difference was found in the load at which AE events began to occur regardless of wet-combination. Further, the shapes of the load-AE curves of the papers were generally similar to each other, i.e., the slopes of the curves were quite low at first, and then gradually and finally steeply increased just before the maximum load. However, the shape of the curve differed somewhat with increasing number of paper plies, i.e., the period of the gradual increase in AE events before the acute occurrence was prolonged for papers made by wet-combination. The longer period indicates an increase

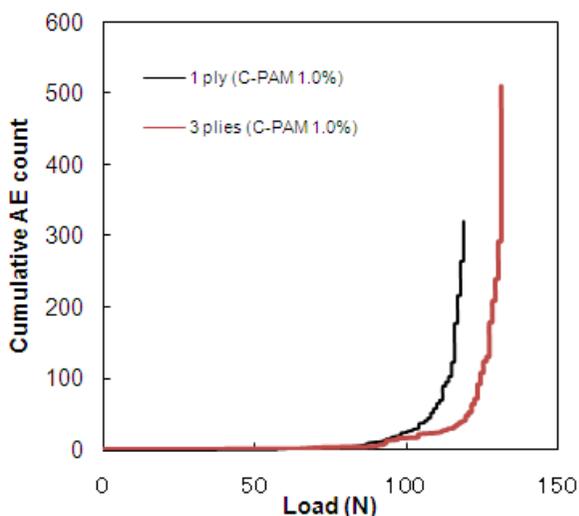


Fig. 3 Representative relationship between the cumulative AE event count and tensile load for the papers containing C-PAM at the total content of 1 %, showing the effect of the number of paper plies.

in the plastic deformation region, leading to higher strain at breakage and an increase in tensile strength. The longer period of gradual increase in AE events (microfailures) could be interpreted as follows: a kind of discontinuity of paper structure caused by wet-combination might reduce a possible development of microfailures leading to sheet failure. Better sheet formation caused by wet-combination could be another reason for the strength increase. However, paper samples in this study were made as the sheet formation kept constant as possible.

#### **4. Conclusions**

Wet-combination with paper plies containing C-PAM brought about an increase in the plastic deformation region, leading to higher strain at breakage. The increase in tensile strength of the wet-combination papers was strongly related to the enlarged strain at breakage rather than the increase in fiber bonding. The existence of a layered structure formed by wet-combination may lengthen the period of gradual increase in microfailures at the plastic deformation region and delay the acute occurrence of microfailures just before the maximum load. Heterogeneous location of a strong paper ply containing C-PAM, especially one side location, decreased the increment of the tensile strength. In contrast, the compressive strength increased with increasing total C-PAM content, regardless of wet-combination. However, the increment was small for papers in which the strong paper ply containing C-PAM located at the center or on one side. Tensile strength in the Z-direction, shown as internal bond strength, was governed by that of the weakest paper ply, and wet-combination brought about a further decrease in the Z-directional tensile strength.

#### **Acknowledgments**

The authors express their huge thanks to Arakawa Chemical, Rengo, and Showa Products Cos. Ltd, for their financial support and to the Laboratory of Wood Processing in the Graduate School of Agriculture, Kyoto Univ for permission to use the AE measurement system.

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(原稿受付 2012年6月27日)

(審査受理 2012年8月27日)

## 紙力増強剤を含むすき合わせ紙の強度的性質

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カチオン性ポリアクリロアミド系紙力増強剤(C-PAM)を加えて強度を増加させた紙をモデル的にすき合わせ、それらの強度的性質に及ぼすすき合わせ、すなわちすき合わせ層数およびC-PAMを加えた高強度の紙層内位置の影響を検討した。その結果すき合わせは、微小破壊が徐々に増大する塑性引張変形域の増加をもたらし、微小破壊が急激に増大して破壊に至るまでの引張破断伸びを増加させ、結果として引張強度を増大させる。ただしC-PAM含有紙層が偏在するとその増分は比較的小さい。他方紙面方向の圧縮強度は、すき合わせの影響をあまり受けずに、単に紙力増強剤量の増加と共に増大する。ただし、C-PAM含有紙層が全紙層の中央あるいは片側に偏在するとその増分は比較的小さい。厚さ方向の引張強度でもある内部結合強度は、強度の低いC-PAM非含有紙層のそれで決まり、さらにすき合わせ面に生じる欠陥によりさらに強度を減少させる。

**キーワード：**紙力増強剤、引張強度、圧縮強度、内部結合強度、すき合わせ、微小破壊、引張破壊伸び、アコースティックエミッション