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# RESEARCH ON ADHESIVE JOINT STRENGTH DEPENDENCY ON LOOSE LEAF POSITION IN A TEXT BLOCK

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Original scientific paper

This paper deals with the binding ability of commercial high grade paper used in a perfect binding process. The adhesive binding strength changes happen as a consequence of adhesive rigid characteristics. Paperback bond strength evaluation is performed by pull-test measurement and visual assessment. Paperback contains a number of the loose leaves' binding edges that are bonded into assembly. The bonded assembly is discussed in terms of adhesion between hot-melt adhesive and paper (substrate). Adhesion strength within the inter phase includes the terms of adhesive joint strength. The research shows that a loose leaf position in a text block has a strong influence on adhesive joint strength changes, although other independent variables (paper and adhesive properties, binding process) have significant impact on the paperback mechanical durability. For this reason measurements of paper properties were included. The adhesive joint strength analysis results were obtained by the statistical regression method. The main goal of the paper is to determine the adhesive joint strenght dependency on a loose leaf position in the text block.

***Keywords:*** *adhesive joint strength, loose leaf binding edge, mechanical interlocking adhesion*

## Istraživanje ovisnosti čvrstoće slijepljenog spoja o položaju pojedinačnog lista papira u knjižnom bloku

Izvorni znanstveni članak

Ovaj rad bavi se problematikom uvezivanja papira, prilikom korištenja "perfect binding" tehnike uvezivanja. Promjena čvrstoće uvezivanja pojavljuje se kao posljedica karakteristične krutosti ljepila. Procjena čvrstoće spoja provedena je mjerenjem sile kidanja pojedinačnog lista papira i vizualnom vrednovanjem. Određeni broj pojedinačnih listova papira su međusobno povezani preko linije veza u meko uvezanu lijepljenu knjigu. O njihovom međusobnom povezivanju raspravlja se kroz termin djelovanja sile adhezije između taljivog ljepila i papira kao podloge. Jakost djelovanja sile adhezije na graničnoj površini izravno se odnosi na čvrstoću slijepljenog spoja. Istraživanja potvrđuju kako položaj pojedinačnog lista papira u knjižnom bloku ima snažan utjecaj na promjenu rezultata čvrstoće slijepljenog spoja, iako ostale nezavisne varijable (svojstava papira i ljepila, proces uvezivanja) imaju značajan utjecaj na mehaničku izdržljivost meko uvezane lijepljene knjige. Stoga su obuhvaćeni rezultati mjerenja svojstava papira. Rezultati analize čvrstoće slijepljenog spoja izvedeni su statističkom regresijskom metodom. Glavna svrha rada je odrediti ovisnost čvrstoće slijepljenog spoja o položaju pojedinačnog lista papira u knjižnom bloku.

***Ključne riječi:*** *čvrstoća slijepljenog spoja, linija veza pojedinačnog lista papira, mehanizam lijepljenja mehaničkim zaključavanjem*

## 1 Introduction

Adhesive perfect-bound product is Paperback, a general binding technique which uses hot-melt adhesive in book binding. Application of the hot-melt adhesive includes book-block spine preparation. Furthermore, adhesive binding quality control includes critical factors, such as paper surface properties and paper grain direction [1]. The adhesive bond strength mainly depends on paper surface properties. Paperback handling is the second most critical quality factor after binding process. The Flat-back binding method includes assembling loose leaves together at binding edge to create a text block [2]. Furthermore, the text block spine is filled with thick layered film of rigid adhesive. The loose leaves are locked in the fixed text block spine without radius. Therefore the Paperback openability is reduced. Paperback handling describes book block quality of adhesive joint strength. Adhesion of the adhesive to adherends and cohesion of adhesive film are main contributors to the loose leaf adhesive joint strength [3]. On the other hand, short times of thermoplastic adhesive setting induce adhesive rigid film on the top of the text block spine. The adhesive rigid characteristics mainly influence the loose leaf adhesive joint strength changes after Paperback opening [4]. These changes are mostly different given the loose leaf position in text block.

In the past, several studies analyzing Paperback adhesive binding strength on different high paper grades were conducted. Leekley et al. [5] studied the relationship between paper properties and Paperback adhesive binding behavior. The study was concerned with determining the critical paper surface tension and importance of its properties for the Paperback adhesive binding strength. Gastel [6] studied loose leaf adhesion joint strength in adhesive binding. He established that loose leaf binding edge characteristics are the most important criterion of Paperback adhesive binding strength. Korhonen [7] studied all factors affecting the Paperback adhesive binding strength. She established process variables of perfect binding techniques due to Paperback volume binding edges roughness characteristics influence on Paperback mechanical durability.

Paperback adhesive binding strength is defined as the force necessary to pull apart the substrates that are bonded together. Adhesion refers to the interaction of the adhesive (hot-melt) and substrate (paper) surface. Mechanical strength is dependent upon chemical bonds in adhesive, substrate and adhesive-substrate interface. Adhesive strength is measurement of failure; the process determines where the localized stress exceeds the bond strength under specific test conditions. The bond strength includes two factors: stress concentration and weakness in surface layers. One concept is the idea of the bonded assembly being a series of links representing each phase with the failure occurring in the weakest link [8].

Generally the adhesive bond is stronger than the substrate so that the failure mechanism is one of substrate fracture mostly on loose leaf binding edge. Also surface preparation provides the adhesive and substrate interaction. Furthermore, material science (surface chemistry and morphology) has a strong influence on the interaction. The paper surface preparation involves mechanical treatment which contributes to adhesive wetting. Furthermore the adhesive's rheological property contributes to adhesive strength. Adhesive solidification describes adhesive film cohesion strength. Bonding process involves short assembly time with pressure. Accordingly adhesive application begins on paper surface. Furthermore the adhesive penetration time is an important factor. The adhesive has to flow over the bulk paper surface into the cavities that are caused by the roughness that is present with almost every surface. Many factors control the surface wetting, including adhesive viscosity and bond line with assembly pressure. Short assembly time explains the adhesive fixed and hardened state. Bond form occurs immediately after the adhesive and the adherend are brought together.

In a mechanical interlocking, adhesive provides strength through reaching into substrate cavities [9]. This adhesion theory excludes the adhesive and substrate physical and chemical interaction. Friction forces prevent detachment of adhesive and substrate. Adhesive wets the substrate including interlocking form. The mechanical interlock size is not defined due to the cavities volume changes. Substrate surface roughness contributes to the energy dissipation around the cavities and in the substrate bulk during tensile force. The stress-strain curve includes the substrate response to tension force. The energy loss is major component of adhesive joint strength.

Adhesive bond strength degradation rate depends on stress-strain and handling conditions. Furthermore adhesive bond strength durability mainly explains adhesive joint strength. The adherend establishes ultimate adhesive joint strength. The physical bonding degree (mechanical interlocking adhesion) with the hot-melt adhesive depends on adherend stability. Therefore adherend characteristics influence bond line damage and the adhesive joint fails.

## 2 Experiment

### 2.1 Raw Materials

Specific paper properties (Tab.1) were measured before proceeding to the adhesive perfect-bound paperback. The paper samples conditioning was performed in compliance with Standard ISO 187, over 24 hours [10]. Paper basic weight was measured according to Standard [11]. In addition, the tensile strength with constant rate of elongation of machine-made paper was tested [12]. The paper furnishes significantly influence adhesive bond. The paper surface roughness and water absorptiveness were determined [13, 14]. Arithmetic mean was calculated based on ten measurements, paper lower (B) and upper (A) side separately. Commercial high paper grade, type Amber graphic uncoated paper 100 g/m$^2$) was tested.

### 2.2 Paperback samples preparation

Ten paperback samples with identical text block spine treatment condition were made according to preliminary test. Optimum adhesive bond includes test adjustment result in order roughening type. The binding process was performed under standard conditions (ISO 187). The ingredients combination, ethylene vinyl acetate copolymer [15] grades and tackifying resin have influence on the adhesive melting point. Adhesive type Planatol HM 6010, temperature 170 °C, viscosity 2965 mPa·s, adhesive film thickness 0,80 mm and adhesive open dry time 600 books/hour (Type: Muller Martini Pony 5) with strong clamping effect were included. Adhesive viscosity was determined in accordance with standard [16] by ISO-Viscometer 550 with Plate Cone. Three measurements were used to calculate adhesive viscosity. The paperback unprinted sample specification is 15 cm (width) × 21 cm (height) × 64 loose leaves (volume). The paper grain direction runs parallel with paperback height. Furthermore, the loose leaves binding edges run parallel with grain direction. The binding edges are filled with rigid adhesive and loose leaves are locked in fixed text block spine without radius, Fig. 1. Arithmetic mean is calculated on the basis of ten measurements, each paperback sample separately.

**Table 1** Specific paper properties

| | Value | ISO Unit |
|---|---|---|
| Basic weight | 93,87 | g/m$^2$ |
| Thickness | 113,64 | μm |
| Ash content | 28,46 | % |
| Calcium carbonate content | 22,36 | % |
| Surface roughness A | 178,8 | ml/min |
| B | 167,5 | ml/min |
| Surface absorptiveness A | 50,7 | g/m$^2$ |
| B | 40,21 | g/m$^2$ |
| Tensile index MD | 53,56 | N·m/g |
| CD | 26,77 | N·m/g |
| Strain rate MD | 1,73 | % |
| CD | 2,75 | % |
| Tensile stress MD | 48,69 | MPa |
| CD | 24,33 | MPa |
| Elastic modulus MD | 4,375 | GPa |
| CD | 2,258 | GPa |

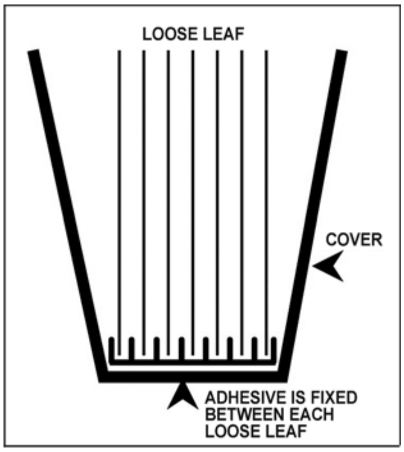



**Figure 1** Paperback - the binding edges filled with rigid adhesive

### 2.3 The adhesive joint strength measurement

The adhesive joint strength is appropriate measure of paperback binding strength. A binding endurance test or pull-test determines the force required to pull a loose leaf from text block spine. The disposition of ten loose leaves in relation to each other is frequently identical (A-J), a test scheme is given in Fig. 2. Its purpose is to determine the adhesive joint strength of the method used to secure the loose leaf. The loose leaf (A-J) is pulled with uniform force along the binding edge.

The paperback is clamped into position by the bottom jaw of the testing device with a loose leaf held in a vertical position by top jaw. The jaws are separated and the force required for tearing the loose leaf or pulling it from bond line or adhesive-adherend layer is measured (Fig. 3). The

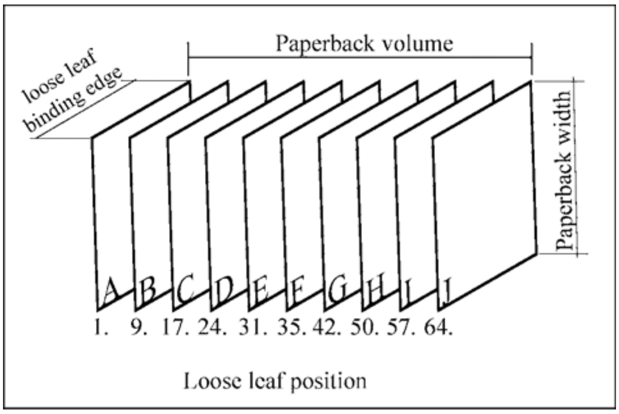



**Figure 2** Paperback - Loose leaf dispositions

total load force is divided by the loose leaf height in centimeter to give the loose leaf pull unit of measurement as N/cm [17, 18]. The result $F_z$ (N/cm) is compared with a rating of adhesive joint strength (bad, sufficient, good and very good binding strength) according to FOGRA guidelines for page pull test [19, 20].

The local tensile stress and tensile strain under given load conditions in adhesive and adherend layer determine adhesive bond strength. Generally, adhesive bond line strength describes loose leaf adhesive joint strength.

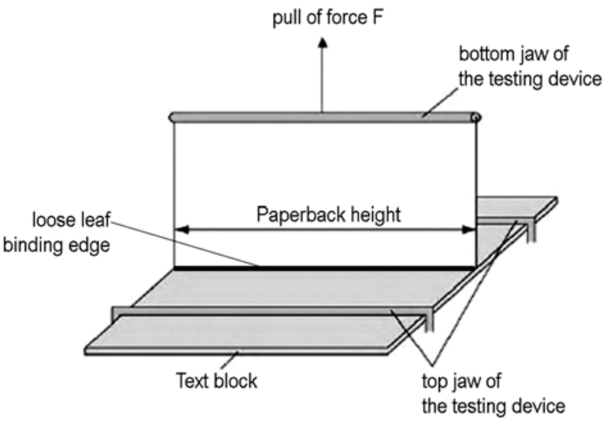



**Figure 3** Adhesive joints strength measurement (pull test device)

The Paperback binding quality was tested by Martini Tester tensile device, Model VA. The pull-test results are represented as the Tensile index (N·m/g), due to comparative paper high grades tensile strength and paperback adhesive binding strength. Text block thickness $d_{\mathrm{TB}}$ (mm) is determined from paperback volume (64 loose leaves) as follows:

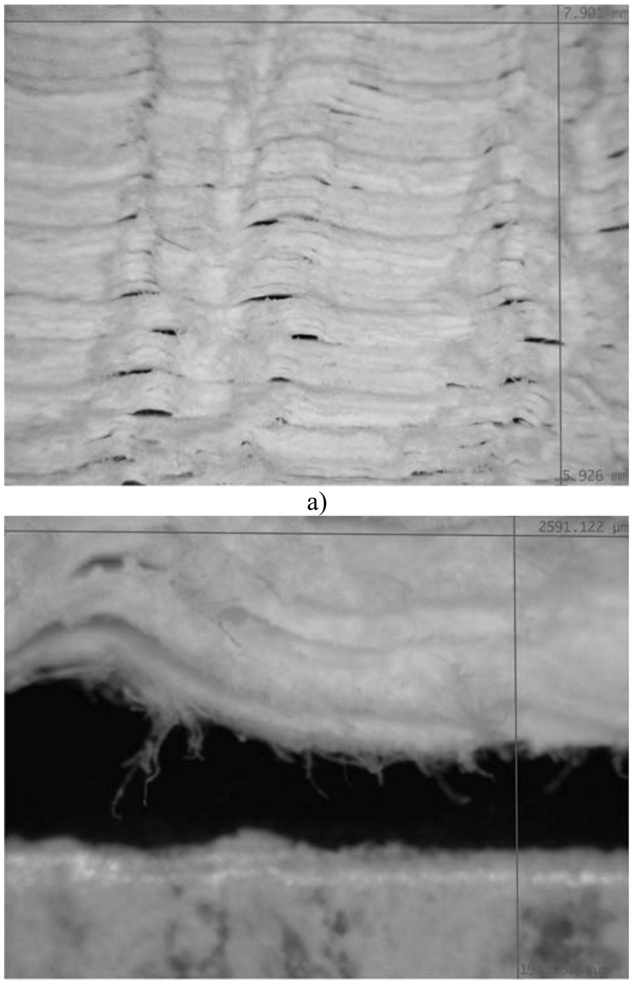
$$d_{\mathrm{TB}} = \sum_{1}^{64} \text{ thickness of the loose } leaf_i\text{, mm} \qquad (1)$$

Text block thickness (mm) is calculated from paper thickness (Tab.1). Furthermore, adhesive joint strength changes appear as a consequence of text block thickness changes. In addition to the loose leaf position (A-J) is manly contributor of changes in paperback adhesive binding strength assessment. Furthermore the loose leaf position impact on adhesive joint strength changes mostly after paperback handling. Therefore the bond line strength changes appear as a consequence of paperback handling.

On the other hand, bond line strength explains adhesive-paper interface intensity as a result of chemical bonds. In addition to loose leaf adhesive joint strength assessment depends on binding edge characteristics. Cavity regular form mainly contributes to mechanical locking adhesion and chemical bonds intensity. Text block spine mechanical treatment involves adhesive joint strength assessment only from adhesion theory aspects.

## 3 Results and discussion

Tests were carried out in two areas: paper parameters testing and Paperback adhesive binding strength testing (Tab. 1 and Fig. 3). Generally uncoated wood free paper contributes to adhesive binding strength most likely due to paper surface roughness and paper basic weight increase (Tab. 1). Furthermore paper mechanical property in cross machine direction as strain rate (Tab. 1) is also the contributor of loose leaf adhesive joint strength. The bond ability increasing can be explained by mechanical interlocking adhesion [] of loose leaves binding edges. The loose leaf contains a large number of regular form cavities after mechanical treatments (Fig. 4a). Adherend was examined in environmental scanning microscope (Fig. 4a-b).

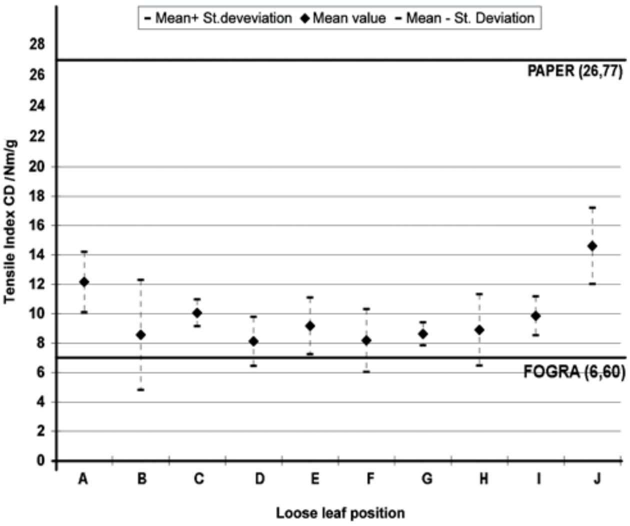


a)

b)

**Figure 4** Paperback volume part - cross section view (A) loose leaves binding edges (magnification 70×; 7,90 × 5,93 mm) after text block spine treatment; (B) A and B loose leaf position - binding edge cross section view (magnification 200×; 2591,12 × 943,32 m)

Furthermore, paper thickness increase is the main contributor of cavity regular form. Therefore higher paper basic weight confirms paper thickness increase. Text block spine surface volume increases due to loose leaf regular cavities form. The surface volume increasing directly explains mechanical interlocking adhesion as higher adhesive penetration rate into loose leaf cavities. The adhesive locks mechanically to the loose leaf binding edge. Mechanical interlocking adhesion is the main contributor to bond strength [22]. Adhesive melt characterization and thermoplastic ingredients influence the cohesive strength of dry adhesive layered film. Furthermore bond strength depends on adhesive right viscosity range according to the process thermoplastic bonding window [23, 24]. Adhesive surface tension, heat adhesiveness initial tack, its opening time and setting time are important parameters for development of cohesion forces in a bonding process [25, 26]. Generally, a good adhesive binding strength result by FOGRA guidelines explains optimal bonding process (Fig. 5). Furthermore significant mechanical interlocking adhesion results from adhesive surface tension. On the other hand the adherend increases mostly contribute to the improvement of adhesive joint strength results. Adherend increasing describes the cavities surface volume which improves adhesive kinetic wettings and its plastic deformation into cavities. The paper surface absorptiveness enhancement leads to adhesive kinetic wetting. Paper no-sizing surfaces and adhesive vinyl acetate component are polar characteristics [27]. Polarity mainly contributes to bond strength. Changes of adhesive joint strength results that depend on loose leaf position can be explained by adhesive inelastic characteristics. Text block spine without radius is main causer of adhesive joint strength reduction after Paperback handling [28]. This fact is related exactly to the middle of text block.




*Mean values were calculated from 10 measurements for each loose leaf

**Figure 5** Adhesive joint strength measurement results

Adhesive joint strength reduction presents loose leaf B, D, E, F, G and H position. Furthermore, all results are significantly higher than FOGRA recommendation. On the other hand the enhancement loose leaf position A and J results are explained by side gluing.

Paper ash content and calcium carbonate content influence the results' decrease. Generally the inorganic components cause mechanical interlocking adhesion reduction. Fig. 6 presents paper surface characteristics after testing. Segment of loose leaf binding edge contains fibers layered structure binds with inorganic components.

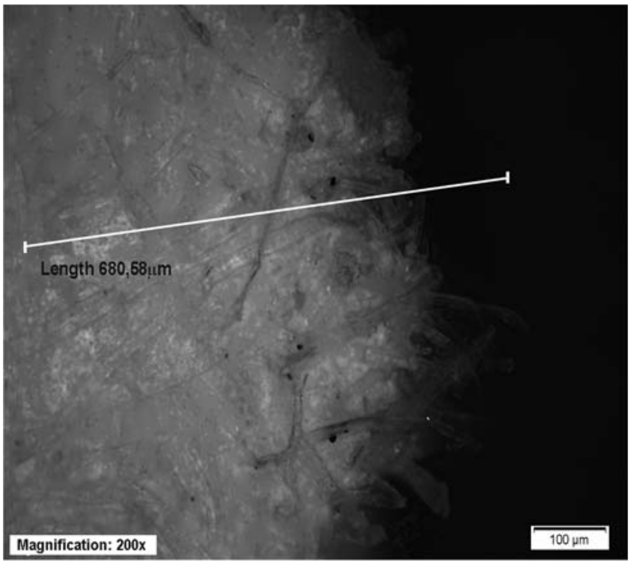



**Figure 6** Paper surface characteristics - loose leaf binding edge after testing

Furthermore, cross section view of loose leaf binding edge confirms adhesive kinetic wetting and inorganic component remaining into cavities after testing (Fig. 7). Free fibers on the top binding edge explain optimal adhesive kinetic wettings. On the other hand, cavity inner part of binding edge consists of inorganic particles without fibers. Generally, photomicrographs under 50-200× magnification show free fibers appear on the top of loose leaf binding edge as a result of mechanical interlocking adhesion.

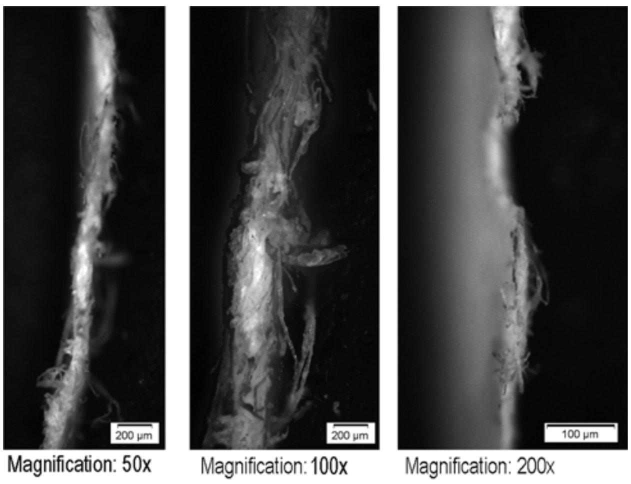



**Figure 7** Loose leaf binding edge after pull testing - cross section view

Fig. 8 presents adhesive joint strength changes through Paperback sample volume (A-J). The adhesive joint strength mean value (Fig. 7) is entirely different with regard to loose leaf individual result (A-J). The adhesive rigid characteristics explain this phenomenon. After Paperback handling, adhesive rigid characteristics partly contribute to adhesive joint strength reduction. Inorganic ingredients (paper dust) rarely remain in cavities and rigid adhesive separates easily from paper fibers layered structure. Generally paper dust appears as a consequence of the text block spine routing principle in perfect binding technique. This binding technique with a rigid adhesive mainly shows the same problems with different high paper grades. The paper chemical properties (increase of ash content) mainly contribute to adhesive locks reduction. Furthermore, the paper mechanical properties mainly contribute to adhesive binding strength. Regression polynomial function of the fourth level verifies this statement. Determination of coefficient result $R^2 = 0,97$ and $p < 0,01$ is indication of regression quality (Fig. 9).

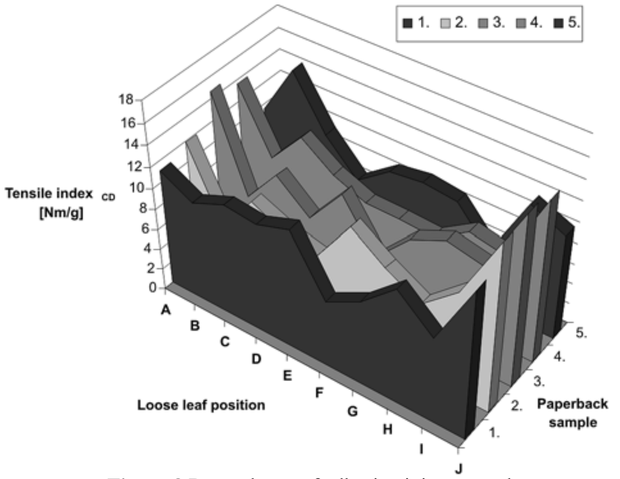



**Figure 8** Dependency of adhesive joint strength and position of loose leaf

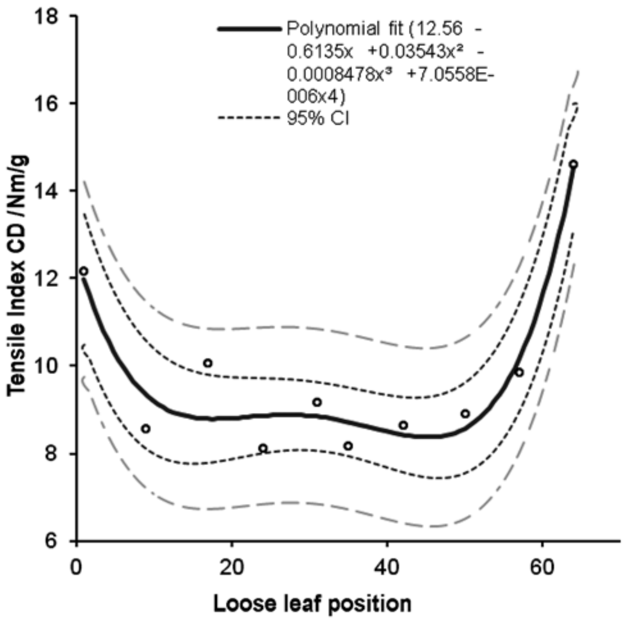



**Figure 9** The relationship between adhesive joint strength and loose leaf position

Paper mechanical properties in lateral direction (CD) are points of interest (Tab. 1). Paper tensile index$_{CD}$ 26,77 N·m/g and strain rate$_{CD}$ 2,75 % result are contributors to adhesive joint strength result. Furthermore, loose leaf binding edge (adherend) tends to stretch. Generally paper viscoelastic behavior has positive impact on bond strength [29, 30]. On the other hand, paper elastic modulus$_{CD}$ result 2,258 GPa confirms the reduction of paper bonding degree. This result explains the paper elastic breaking strain through adhesive-paper interface, fibers network bonds accurately. Photomicrograph confirms that fact (Fig. 7).

The paper extension is the main cause of adhesive joint strength. The external load is transmitted across adhesive-paper interface fiber layered structure. The paper interface bond fiber failure determination is pointless due to loose leaf binding edge characteristics. In other word, the loose leaf binding edges internal resistance results in the deformation or pull out from text block spine.

The full understanding of Paperback adhesive binding strength includes the mode of loose leaf failure characterization. This binding technique recognizes three typical characterizations: cohesive, adhesive joint and substrate failure. Visual assessment tests show that loose leaves lead to a higher adhesive joint strength, the failure of adhesive joint (adhesive-paper interface) is mainly confirmed (Fig. 10). Furthermore, loose leaf position A and J confirmed substrate failure due to side gluing. In addition to loose leaf position E (middle of paperback volume) had mixed adhesive joint/substrate failure. The substrate fracture outside of loose leaf binding edge leads to adhesive out of locks after paperback handling.

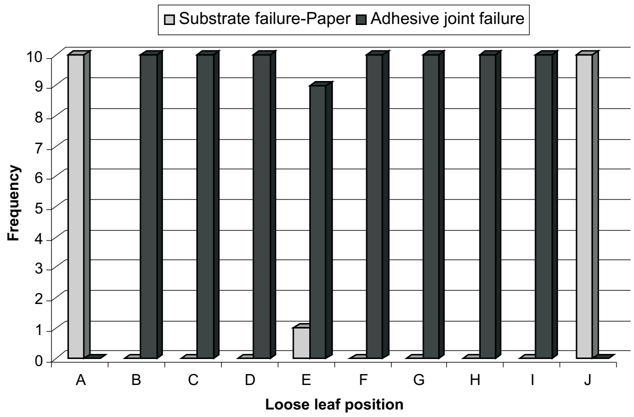



**Figure 10** Visual assessment – Paperback failure characterization

A) Substrate failure - loose leaf position A (binding edge)

B) Substrate failure - loose leaf position J (binding edge)

C) Substrate failure - loose leaf position E (outside of binding edge)

D) Adhesive joint failure (adhesive/adherend interface) loose leaf position B-D and F-I

**Figure 11** Paperback failure characterisation photographs. Photomicrographs under 200× magnification, showing loose leaf binding edge after testing

Inspecting paper through a microscope is perhaps the appropriate way to understand the nature of adhesive-paper interface (Fig. 11A-D). Paper fiber layered network runs parallel with loose leaves binding edge (Fig. 11D). Adhesive joint failure explaines as fibers network pulls out from adhesive rigid film interface. Furthermore, fibers network contributes to bond strength due to the fibers' better connectivity with rigid adhesive. Generally the paper thickness increase is the main cause of bond strength through fiber network interface. Paper fibers layered network contributes to mechanical interlocking adhesion and adhesive bond strength. On the other hand, substrate failure (Fig. 11A-B) appears due to perfect binding technological process (side gluing). Adhesive remains on loose leaf binding edge and loose leaf is damaged partially after testing.

## 4 Conclusion

This study focuses on the binding ability of uncoated wood free high paper grade using the perfect binding technique with hot melt adhesive rigid characteristics. The Paperback adhesive binding strength determination included pull-test measurement and loose leaf visual assessment. Paper mechanical properties in lateral direction and intensity of mechanical interlocking adhesion are main contributors to adhesive joint strength. On the other hand, adhesive rigid characteristics cause reduction of adhesive joint strength in the middle of text block after Paperback opening. This explains result changes dependency on its position in text block. This statement is verified by regression polynomial function of fourth level. Furthermore, adhesive polarity and cavities regular form in text block spine improve bond strength. Generally, paper surface roughness (paper, text block spine) directly contributes to enhancement of adhesive kinetic wetting and adhesive joint strength. Inorganic ingredient is the main cause of the bond strength reduction. This problem exists as a consequence of perfect binding technique; paper dust appears after spine treatment. Further it is concluded that uncoated wood free paper shows excellent results by FOGRA guidelines recommendation. Visual assessment directly confirms this fact given the adhesive joint failure characterization. Photomicrographs exactly show failure position and mechanical interlocking adhesion intensity. Accordingly, paper fiber network layered structure interface with rigid film layer adhesive mostly contribute to the loose leaf adhesive joint strength.

These preliminary results can be used in future. Perfect binding technique optimization will include uncoated paper of different paper thickness and adhesive thickness film with same adhesion surface tension, heat adhesiveness initial tack, open and setting adhesive time.

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