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BACTERIAL NANOCELLULOSE ENHANCED CARDBOARD ADHESION JOINT TESTED WITH Y-PEEL AND T-PEEL TESTING METHODS

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Abstract: *The demands on the adhesive joints for packaging are important during conversion, transport, storage, shelf exposure, and end-of-life operations of paper-based packaging. During this lifecycle, the adhesive joint is under constant loading from environmental conditions (e.g. forces from the surroundings such as other packaging units, dynamical forces from the machines, and/or climate changes such as temperature and relative humidity). The contribution from mechanical bonding can be particularly important for solid boards where the adhesive bonding is achieved through the mechanical interlocking of the adhesive into irregularities and pores of the carton-board surface. The formation of an adhesive joint with dispersion adhesives is affected by the rheological properties of the adhesive layer, the structure and absorption properties of the board surface. There are many test methods intended to determine the strength of an adhesive including peel, shear, cleavage and tension tests. Peel tests are common for tapes, labels, coatings and other bonded materials. The most common methods include T peel, Y peel and angle peel tests using tensile test machines. The cardboard adhesives that are currently in use have a large share of dispersion adhesives (PVAC, EVA, acrylic and PU) which are water-based adhesive systems that form bonds through physical hardening when the water evaporates. Sustainability issues require replacement of these chemicals with bio-based ones. Based on our previous research, bacterial nanocellulose (BNC) can increase the bonding strength of adhesive joints for different kinds of materials. In this study, 3 different cardboards with dispersion adhesive which had the addition of 7% of BNC were used. Zwick multitester with two methods (Y peel and T peel) was used to determine the strength of the cardboard joints. The results indicate differences in force elongation results due to different testing methods, where some of the cardboard properties and BNC had a positive effect on the overall adhesion joint strength.*

Key words: cardboard adhesion, bacterial nanocellulose, Y peel, T peel

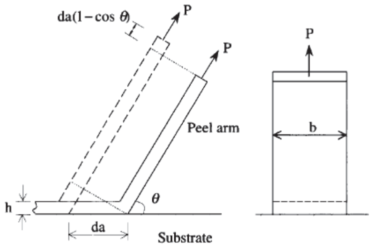
1. INTRODUCTION

Gluing is a very important process in the conversion of coated paperboard packaging products, that is used to join paperboard surfaces together, providing a permanent join. It is also used to erect and close cartons and to provide several functions to graphical products. For a predictable and reliable way to adhere the paperboards is by careful choice of the surface sizing system, interlaminar strength and pigment coating. Gluability is especially important for high packaging line efficiency and runnability. The glue seam must withstand forces arising from the package contents during the lifetime of the package. The established way to assess gluability is to examine the tear behaviour of a glue seam between a pigment-coated surface and, usually, the reverse side of a carton flap. When a barrier effect is created at the surface of either substrate, it slows down the dehydration rate of the glue considerably thereby reducing the tendency to tear fibre upon separation. The main criterion in determining successful gluing of packaging products is almost always 100% fibre tear upon separation of the glue joint. Failure to achieve fibre tear along the glue joint is considered a failure in packaging conversion. In a recent study by Dohr and Hirn (2021) different paper properties of the adhesive strength of starch gluing were investigated with surface roughness, wettability and glue penetration measurements. The results suggest that the roughness (macro) played a minor part while the main parameters were fibre wetting and penetration of the glue into the fibres.

Nanocelluloses (i.e., bacterial nanocellulose, cellulose nanocrystals, and cellulose nanofibrils) are cellulose-based materials with at least one dimension in the nanoscale. These materials have unique and useful properties and have been shown to assemble at oil-water interfaces and impart new functionality to emulsion and latex systems (Kedzior, Marway & Cranston, 2020). Cellulose fibres on the nanoscale are divided in four groups: (1) bacterial cellulose nanofibers, (2) cellulose nanofibers prepared by electrospinning, (3) nano fibrillated cellulose plant cell fibres and (4) nanorods or cellulose whiskers. Processing techniques have a significant impact on the adhesion properties of the resulting cellulose nanofibers in composite material applications. Bacterial cellulose structure is more homogeneous than standard cellulose. This property together with a lack of irregularities lead to both superior reinforcement and thermal expansion properties when used with matrix materials. The nanocellulose inter- and intra-molecular binding are accomplished through hydrogen bonding. Bacterial cellulose fibres have normally a degree of polymerization between 2000 and 6000 (Iguchi, Yamanaka, Budhiono, 2000). This relatively low degree of polymerization may limit the adhesion through interpenetrating networks or mechanical interlocking and, for the most part, the adhesion in composite materials is limited to hydrogen bonding through other mechanisms of adhesion that need to be explored.

An overview by Li et al. (Li et al., 2021) based on previous research concluded that the addition of nanocellulose in the pulp, that the surface of NC which is rich with free hydroxyl groups, will be distributed in the gaps between the fibres or on the fibre surface. Nanocellulose is closely combined with the pulp fibres, and therefore it strengthen the adhesion between the fibres, fill the voids in the paper, and achieve the effect of improving the strength of the paper. The research (Kedzior et al., 2020) cellulose and nanocellulose surface issues regarding adhesion conclude that in heterogeneous water-based polymer systems, when BNC, CNCs, and CNFs have been incorporated either by blending with the dispersion of polymer particles post-synthesis, or by adding nanocellulose at the beginning of the polymerization reaction, i.e., in situ incorporation, several interactions occur. The interactions between nanocellulose and monomer droplets/polymer particles are governed by hydrogen bonding, van der Waals forces, surface activity, hydrophobic interactions (if the nanocellulose is surface-modified for compatibility), and they may even be tethered to each other if the polymerization initiator induces some free radical reaction sites on the nanocellulose (Dastjerdi, Cranston & Dube, 2018; Ghosh, Dev & Samanta, 1995; Misra, Mehta & Ketarpal, 1984; Zhou et al., 2011). Due to hydrophilic nature of unmodified nanocellulose, it is generally inferred to be in the aqueous phase (i.e., outside of/between the polymer particles) when blended with latexes or in situ polymerized in the presence of a noninteracting surfactant (that stabilizes the emulsion polymerization) (Kedzior, Marway & Cranston, 2017). As such, the location of nanocellulose relative to the polymer particles and ultimately, their influence on product properties, is a fine balance of the interactions between all components in the polymerization reaction. Despite deep literature review, we have not found a great deal of publication on the topic of using nanocellulose addition in cardboard adhesives as they cover mostly adhesives for wood panels which due to differences in the surface structure have a somewhat different locking mechanism. For example, a recent study of adding nanocellulose components to PVAc adhesives used in cardboard joint gluing showed that replacing parts of the adhesive gave mixed results regarding the z-tensile strength, but almost all concentrations (0.5, 1 and 2 %) were beneficial for the T peel adhesion tests at the room temperature as the average peel force (N/mm) increased for all samples.

The adhesive bonding in general can fail with three different types of failing mechanisms: adhesive failure, cohesive failure or substrate failure. Adhesive failure is mainly caused by improper penetration of the adhesive onto the surface, the substrate failure is connected with the internal bonding of the substrate (in paper packaging the interlocking of fibres and additives or coatings) and the cohesive failure is connected with the insufficient cohesive forces in the adhesive or coating layer itself. To test these failures different kinds of the tests have been developed. The traditional way of analysing the mechanical strength of a hot-melt adhesive joint in the converting industry is to look at the fracture surfaces after manual peeling. The joint is acceptable if there are more than 50% fibres on the fractured adhesive surface; otherwise, it is considered as bad. However, this method is very subjective and based on personal skills and experience (Korin et al., 2007). Peel tests are the most used type of tests for determining adhesion for thin ductile materials because the tests are fast and easy to make and the test rig is simple. The theory behind peel tests is easily derived when the simplifications that the peel arm is infinitely stiff when pulled and flexible in the bending which is made, the total energy to separate the surfaces for these assumptions is known as $G^{\infty E}$. To calculate the energy required for separation, the parameters shown in Figure 1. must be known.



θ $= angle$
P $= force$
$da = separated\ lenght$
b $= width$
h $= height$

Figure 1: Basic peel testing mechanism (Kinloch & Williams, 2002)

The energy is given by the work done by the force (Eq. 1). The distance of the force is da(1 - cos θ) which gives the energy P × da(1 - cos θ) that is required to separate the area b × da, (Kinloch & Williams, 2002).

$$G^{\infty E} = P \cdot \frac{da(1-\cos\theta)}{b \cdot da} = \frac{P}{b}(1-\cos\theta) \tag{1}$$

This is a very simplified model but it is the basic concept for the peel tests. Effects that come into account when a more advanced model is used are elongation of the peeling arm, bending and plasticity in the peel front, kinetic energy and root rotation (the angle in the peel front) (Kinloch & Williams, 2002).

Different kinds of peel tests have been introduced for fibre-based packaging materials like the T-peel test (Edin, Ödberg & Sterte, 2002), the angle peel test (Kinloch, Lau & Williams, 1994) and Y-peel tests (Korin et al., 2007). In the T-peel test method, the adhesive joint is not fixed relative to the tail and the tail is free to move during the test, while in the other two methods no arbitrary movement is allowed. The Y-peel method is a redesign of the constrained T-peel test.

To completely cover all aspects of the substrate and adhesive failures, a list of an extended set of tests is needed like the surface wettability test, PPS Roughness test, K&N ink stain test, water interference method, SEM microscopy, z-directional or internal bond testing which are covering the substrate side of the adhesion system. In our research, we have used the Bendtsen roughness and porosity for the cardboard surface morphology detection, penetration dynamics analyser for water penetration measurements and contact angle measurements for the wettability of the surfaces. For peel testing we have used two methods: T and Y peel testing and we have calculated the forces of the adhesive joint failure.

2. METHODS AND MATERIALS

In this research three commercial coated cardboards (Sample 3 - MM Excellent TOP 230 g/m^2, Sample 2 - MM Grafopak 250 g/m^2, Sample 1 - Kromopak 230g/m^2) were tested with adhesive EUKALIN 6550-VL-80 water-based dispersion (REF samples) (Brookfield viscosity of 1000 mPas). The adhesive was enhanced with a 7% BNC solution (BNC samples) wich was obtained from an alternative raw material – vinegar mother as described in an article by Lavrič, Medvešček and Skočaj (Lavrič, Medvešček and Skočaj, 2020). The surface roughness of the substrates was evaluated according to ISO 8791-2:2013 standard, and the porosity according to ISO 5636-3:2013. The dynamic contact angle (the wettability of the samples) was evaluated with the Fibrodat 1100 measuring device (Fibro System AB, Gustafs, Sweden) using distilled water drops of 4 µl following Tappi T 588 standard. We have used also ultrasound dynamic penetration measurements using the Emtec PDA measurement device.

The samples were prepared according to the ASTM standard D1876 for the T peel testing and the amount of glue was 0.4 g per sample. To ensure even pressure on the whole surface the samples were pressed with a standardized FINAT 2 kg roller. The measurements of the peel test were done on Zwick multi testing machines using the 10 KN measuring head and adjusted specimen holders.

3. RESULTS

The measurement of the roughness and porosity with the airflow-based method is a standard way of the surface morphology and porosity of different cardboards. The measured results presented in Figure 2. show that there are quite large differences in the coated and uncoated sides of the used cardboards. As the adhesive penetrates both sides when a box is glued on the production line the differences in these parameters can indicate changes in the peel adhesion force. Sample 2 had the largest coated side roughness with a value of 480.55 ml/min and the uncoated side had 2491.9 ml/min. The Bendtsen porosity (Figure 3.) shows that sample 2 again was different from the other two samples, where sample 2 both on the coated and uncoated sample sides had low air permeability (3.54 ml/min for the coated and 2.26 ml/min for the uncoated sides), while sample 1 had the highest air permeability.

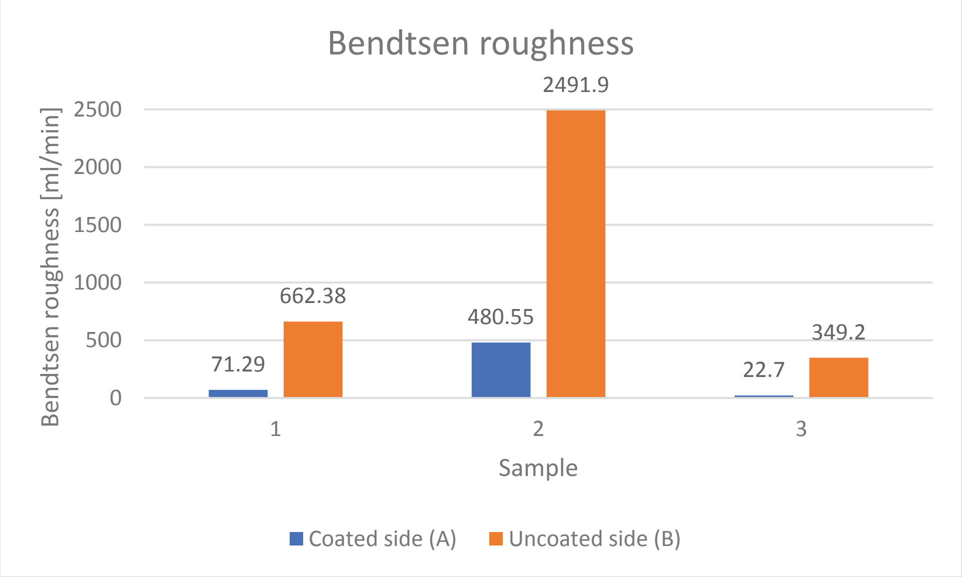


Figure 2: Bendtsen roughness of the sample cardboards

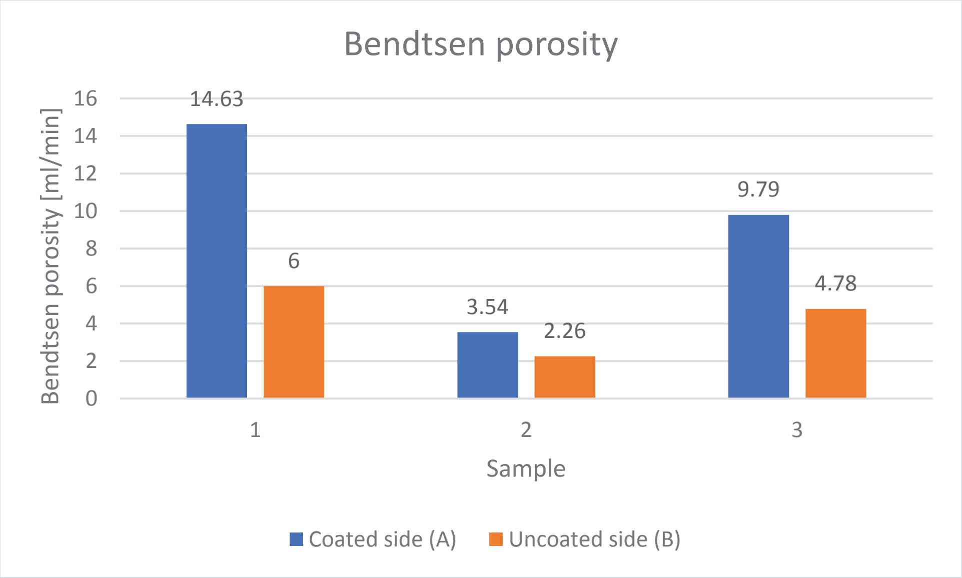


Figure 3: Bendtsen porosity of the sample cardboards

As air and liquids penetrate the surface in different ways we have also tested the samples with the ultrasound method for determining the dynamic penetration of liquids. For the measurements, the Emtec PDA measurement device was used with water and water as the working fluid. Signal intensity decline which indicates the penetration mechanism into the fibrous substrate was measured during 1 minute time. From the absorption curves in Figure 4. one can observe that the surface is quite closed as almost no liquid penetration is happening in the first 2 seconds of the samples exposed to water, except for the coated side of sample 1, which also is similar to the air porosity results. On the other hand for sample 2 (both coated and uncoated)

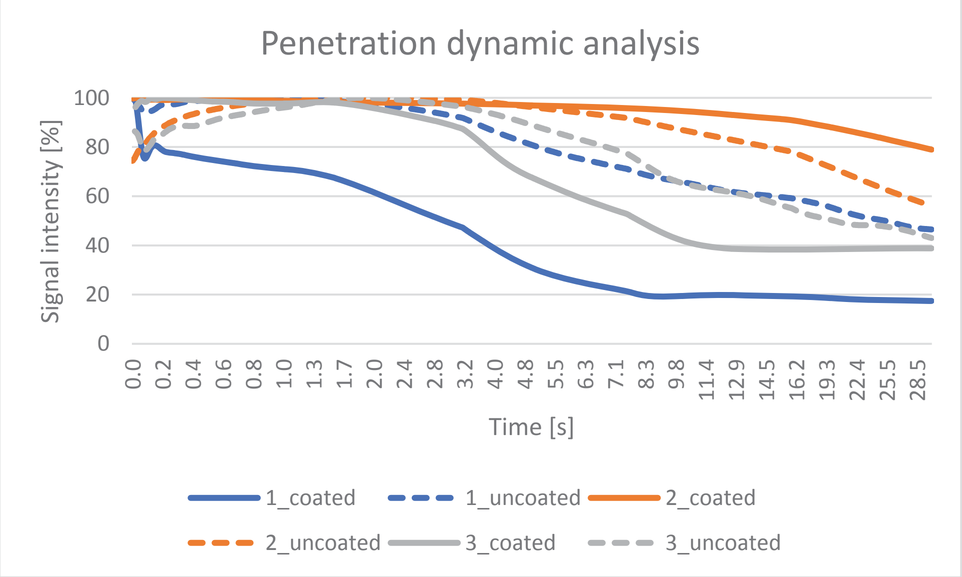


Figure 4: Penetration dynamic analysis of samples with water

The measurement of the dynamic contact angle is presented in Figure 5.

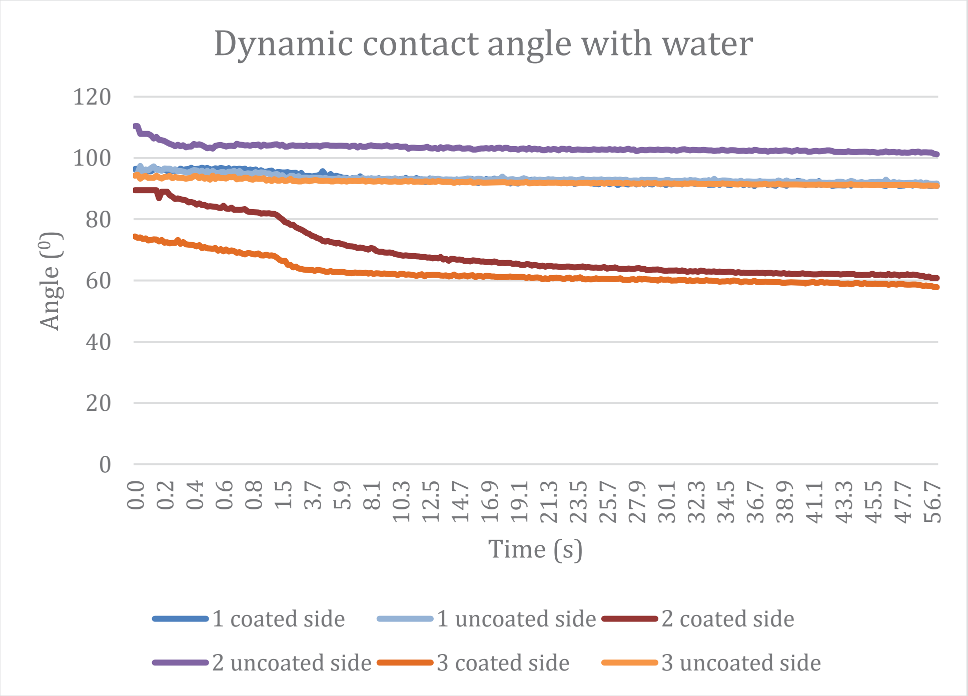


Figure 5: Contact angle measurement of the samples

Before mentioned properties were confirmed again. Sample 2 had the largest contact angle values on the uncoated side and lower value for the coated side (at 1 s 104^0 for the uncoated side of sample 2 and 81.94^0 for the coated side). Sample 1 had the most similar values for both sides at 1s (95.75^0 and 94.95^0) while sample 3 had quite a low contact angle of 68.12^0 for the coated side and 92.93^0 for the uncoated side.

The peel tests were performed with the same speed 254 mm/min. and with a minimal force of 0.1 N. The clamps were also adjusted so the glued cardboard samples were aligned at the same measuring distance. The results of the Y peel are presented in Figure 6 and 7, while the results for the T peel testing is presented in Figure 8 and 9.

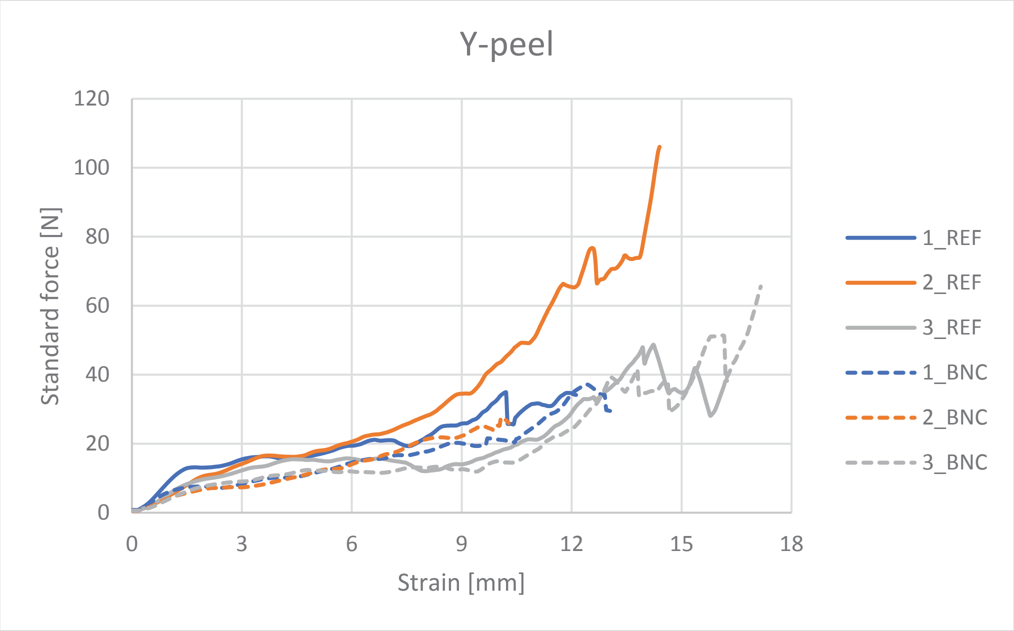


Figure 6: The standard force and strain of the measured samples with the Y peel method

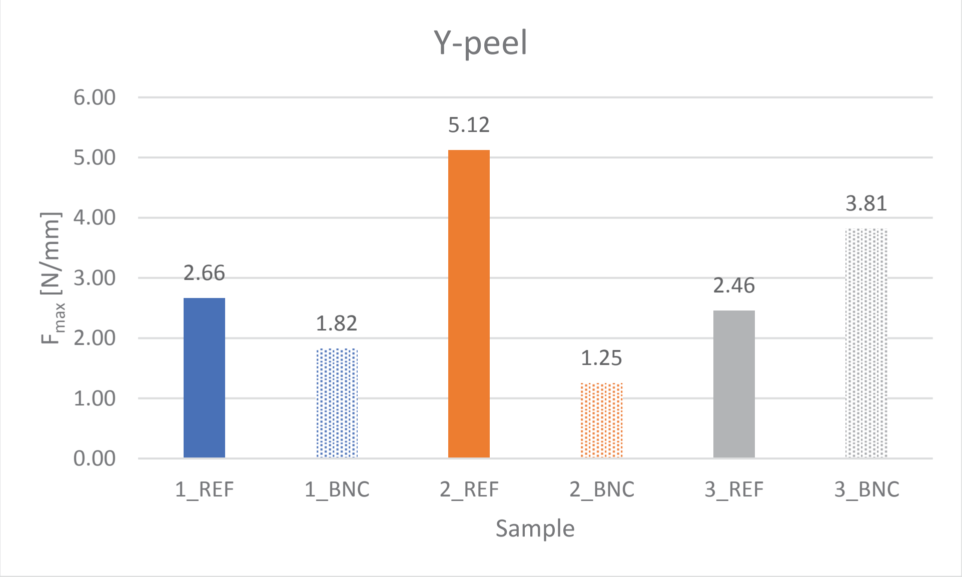


Figure 7: The max force of the measured samples with the Y peel method

From the measurement with the Y peel method, we can observe that the 2_REF sample had the largest force measured. In parallel it was observed, that the same cardboard had the roughest and closed surface and the smallest joint adhesion with the use of BNC. For other samples, the adhesion strength was slightly lower for sample 1 (2.66 N/mm to 1.82 N/mm), but a little bit higher for sample 3 (3.81 N/mm to 2.46 N/mm). It should be noted that for all samples the internal bonding between plies of the cardboard was the main cause of the adhesion joint failure, and the crack propagation was visible in the form of fibre tear. For the T peel testing the results showed some similarities, but opposite results for sample 2.

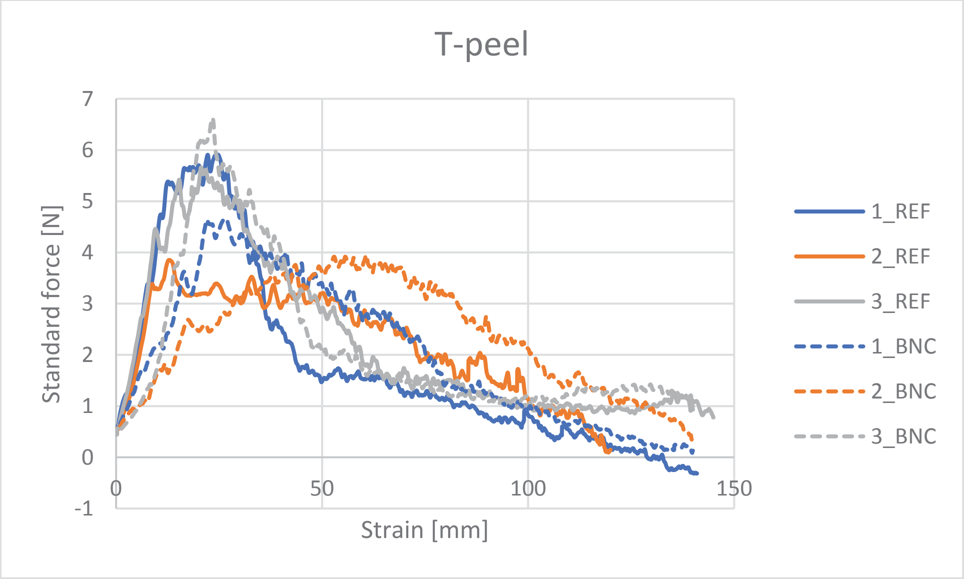


Figure 8: The standard force and strain of the measured samples with the T peel method

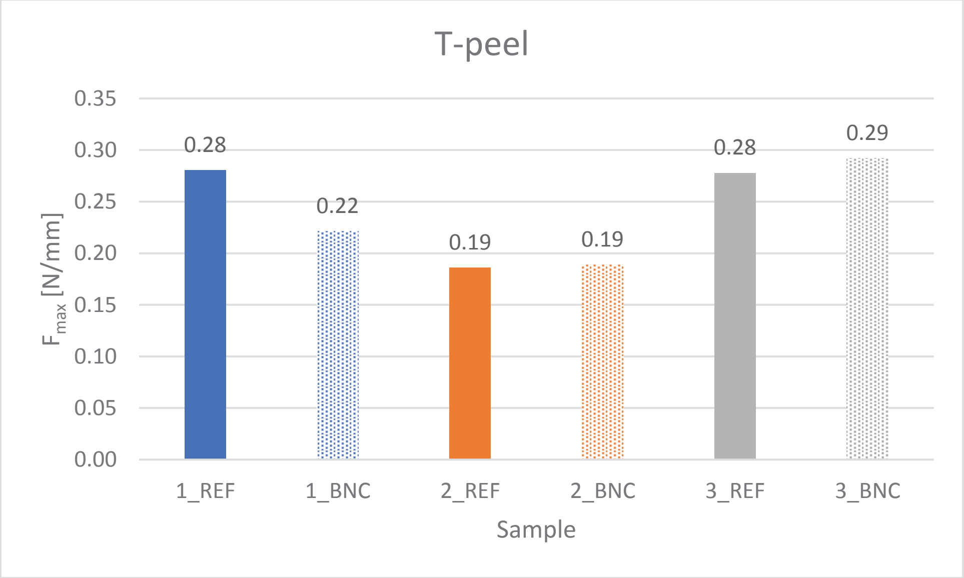


Figure 9: The max force of the measured samples with the T peel method

Sample 1 and sample 4 had similar trends with lower Fmax values in N. For sample 2 this method of peel testing showed no differences but is different from the Y peel method for the reference sample (with no applied BNC). As shown on Figure 8 which indicates larger forces for samples 1 and 3 both for BNC and without which means that the forces applied to the adhesion joint were acting more quickly than in sample 2. As sample 2 had a closed surface and low wettability this can indicate that in the T peel testing the thinner layer of the glue had less elasticity and internal cohesion and the strain on the fibre to coating bonding exerted larger delamination forces. At Y peel as most probably the adhesive stayed on the surface between the 2 plies of the 2_REF sample and the method uses lower angle where the forces have potential effects. As this sample had no BNC (which contains a certain amount of water which can dissolve some of the bonding the fibre network structure and was quite closed regarding the penetrable surface (less chance for mechanical interlocking) the high adhesion separation force is due to strong internal cohesion of the polymer adhesive.

4. CONCLUSIONS

This research tried to indicate some challenges when testing new bio-based materials for use in packaging applications (adhesion joint failure testing). The result indicates that the use of bio-based BNC as a substituting agent of 7% in the water-based polymer matrix can improve slightly the adhesion joint. This value was determined with previous pre-tests, for further values additional testing of BNC compatibility with different water-based polymer solutions needs first to be tested. On the second hand, with the use of different peel tests, one must know the difference in basic cardboard parameters. Our results show that for example the large differences in surface roughness and porosity as well penetration ability are influencing factors if the two peel methods are to be compared. In either Y peel and T peel testing wettability properties as previously tested with other adhesives can be more useful than surface roughness parameters (especially macro roughness). As almost in all samples the fibre tear occurred between the fibre matrix and the surface coating on the coated side of the cardboards additional information needs to be obtained for the internal bonding strength or the z-tensile strength of the base materials. This would show the potential weak points of the fibre layering cohesion failures with the less shallow or deeper penetration of the adhesive and the BNC entangling of the upper layers of this fibre matrix. Further studies will be carried out also on the potential of using also other peel testing angles and the forces applied on the adhesive joints of packaging materials.

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