# UTJECAJ NALJEPNICA NA PROCES RECIKLIRANJA PAPIRA

# THE INFLUENCE OF LABELS ON PAPER RECYCLING

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## Abstract

The growing focus on sustainable products and packaging, raises the question about labels recyclability. The recyclability of labels actually depends on the composition of the labels and their purpose, while the great variety in their creation poses a challenge to their recycling. In this paper, the influence of the polymer labels presence on paper recycling processes is analyzed. Used labels consist of a thin paper backing, glue (adhesive) and polymer with a print. Recycling was performed by chemical deinking flotation, and removal of sticky particles by Somerville recycling test. The efficiency of the deinking flotation process was evaluated by measuring the optical characteristics of recycled paper. The results show that label recycling is difficult but possible, only because the prints on the polymer substrate show a hydrophobic nature, despite the fact that during disintegration they do not break into smaller particles but remain as large specks. Prints together with the polymer substrate are separated during deinking flotation in the form of flotation foam. The problem in label recycling is due to the presence of adhesives .



***Keywords****: Labels, deinking flotation, recycling, optical characteristics, Sommerville test .*

## Sažetak

Sve veći fokus na održive proizvode i ambalažu, dovodi do pitanja mogu li se naljepnice reciklirati? Mogućnost recikliranja njaljepnica zapravo ovisi o samom sastavu naljepnice i njihovoj namjeni a velika raznolikost u njihovom kreiranju predstavljaju izazov njihovom recikliranju. U ovom radu analiziran je utjecaj prisustva polimernih naljepnica na postupke recikliranja papira. Naljepnice se sastoje od tanke papirne podloge, ljepila (adheziva) i polimera s otiskom. Provedeno je recikliranje procesom kemijske deinking flotacije, te uklanjanja ljepljivih čestica Somerville testom recikličnosti. Učinkovitost procesa deinking flotacije evaluirana je mjerenjem optičkih karakteristika. Rezultati pokazuju da se recikliranje naljepnica odvija otežano ali je moguće, samo zbog toga što otisci na polimernoj podlozi pokazuju hidrofobnu prirodu, bez obzira što se tijekom dezintegracije ne lome na manje čestice nego ostaju velikih površina. Otisci se zajedno s polimernom podlogom izdvajaju tijekom *deinking* flotacije u obliku pjene. Problem u recikliranju naljepnica stvaraju prisutni adhezivi, tj ljepila.

***Ključne riječi:*** *Naljepnice, deinking flotacija, recikliranje, optičke karakteristike, Sommervile test.*

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## 1. INTRODUCTION

The printing industry is changing rapidly. According to Smithers, by 2030 packaging will account for almost two thirds of the global print market, while publishing will make up nearly 40% of print volume in 2030, but will still be the biggest loser in the market and will reflected the market for printing substrates [1]. Additionally, according to Smithers, the global demand for printed labels and sleeves will strongly grow and will reach $49.9 billion in 2024, which leads to adjustment of printing equipment builders, ink suppliers and print service providers and shifting end-use demands and the challenge of leveraging the latest technology effectively. These include the further penetration of high productivity printing machines, namely inkjet in production of labels, high quality labels as weel as the need for enhanced sustainability in labelling production [2]. When designing an environmentally friendly product, all product aspects must be taken into account – from its maufucture to its end of life. When designing a packaging material, or label, different materials can be involved into its production, i.e. different materials will form a unified unity. But sustainability doesn't exist in isolation, thus for packaging to be truly sustainable every single part must be eco-friendly [3]. In the formation of sustainable self-adhesive, the most importan parameter are adhesive, materials for production of label, inks, and support on whic the label is placed. All components are equally important in the production od quality product, but it is also important to empasize that all play an importan role in sustainability. In order to increase their sustainability, natural resources instead of synthtetic should be use. Altogether with using natural resources, environmentally friendly process technologies and end-of life solutions are required. It is well known that multi-layered materials are generally difficult to recover and recycle. Thus, design for revlaorization and design for demateralization should be taken into account when lalbels are produced. When creating a sustainable self adhesive label, an LCA method can be useful tool [4]. There is a lot of innovation in the field of eco-labels available on the market today and it is still developing.

Novel technologies and materials change the world as a better place, but simultaneously they can bring new risks and new challenges [5]. Additionally, the increased demand and increased production of labels, leads to the fact that over a period of time there may be a greater accumulation of them as waste material. Also, different combinations of labels and paper (as lalbes backing for example), can lead to their appearance in the paper value chain and can affect the quality of paper recycling. On the other hand, this should be considered from another point of view, as an option for isolation of cellulose fibers from one growing source of waste. Due to the Precautionary Principle, all materials and technologies should be examined in the terms of their potential risks. There is a lack of papers referenced in WoS, Scopus and Google Scholar regrading the possibility of self-adhesive labels on paper recycling. Some of the research are related to the influence of smart labels on paper recycling process [6], [7] wile some of them are related to the environmental evaluation of smart labels [8] or recycling of printed polypropylene (PP) labels with polymer recycling technologies [9]. Thus, this paper evaluates the possibility of using paper with self adhesive labels in paper recycling process and evalutes potential problems.

The amount of non-paper components in recovered paper are depandant upon the collection and handling of paper for recycling. Generally, the non-paper components do not affect recyclability, unless the substances belong to “prohibited materials” (such as (laminated covers, staples, adhesive, polymeric components, varnishes, inks) [10]. An increased use of non-fibre components can lead to increased difficulties in the later stages of the recycling processing chain [11]. Aliaga et al., (2015) concluded that smart labels make no significant changes in mechanical and optical properties of the recycled paper and consequently do not affect the quality of recycled paper [6]. Gregor-Svetec et al., (2016) showed that the presence of printed conductive ink has some influence on deinkability of printed label papers [7]. Adhesives are the essential part of the labels, used for joining materials together. As some other non-paper components (resins from wood, coating binders, ink binders, coatings, impregnation, adhesives) they are the source of sticky impurities. Stickies may cause disturbances in recycled paper production and thus should be removed at the highest degree possible to avoid quality defects in the produced papers (specks, holes) and production process faults (e.g. wet web breaking) [12].

## 2. EXPERIMENTAL

### 2.1. Materials

This paper examines the efficiency of the deinking flotation process of paper with printed self-adhesive labels. The prints were made on a polymer surface glued to paper (Figure 1). The labels were printed in black and yellow (Figure 1.) For each colour, the same printing substrate (polymer) was used. Printing was conducted by Roland VersaUV–LEC 300 printer.



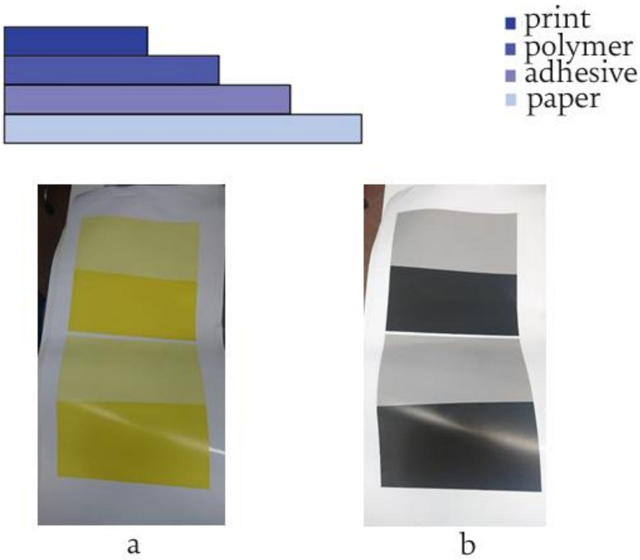


**Fig. 1: Samples - (a) printed yellow and (b) black label**

### 2.2. Methods

Papers with labels were recycled by means of chemical deinking flotation under laboratory conditions. The methods used for recycling are INGEDE 11 recycling method, and INGEDE 11 recycling method in combination with ultrasound (modified INGEDE 11). The process flow of the INGEDE 11 method and modified INGEDE 11 can be seen in Figure

2. As the recycling of labels, which contain large amounts of adhesive causes particle adhesion and formation of stickies, the ultrasound was added to the INGEDE 11 process (modifed process) to see its potential role in the reduction of stickes formation and adhesion of particles to each other, during recycling. During the recycling process, laboratory sheets (handsheets) were made of paper pulp in the stages before and after deinking flotation process.

Additionally, the Somerville test was conducted. This test can be used for the removal of sticky particles and for the separation of sticky particles, dyes, adhesives, and other impurities that can be found in packaging (added during packaging production or gained during its lifecycle) intended for recycling. 25 g of samples were cut into squares (2x2 cm), transferred into a laboratory disintegrator (Enrico Toniolo) filled with 1.5 L 40 °C tap water and pulped for 20 min. The pulp suspension was afterwards subjected to 7 min Somerville screening (TLS Screen Fractionator, SOMERVILLE type – Model SF-10) following a procedure described in TAPPI T 275 sp-12 Standard. All the "rejects" (material separated by the screen) and the "accepts" (fibrous material passing through the screen and possible filler particles) were collected, filtered and dried overnight to obtain the solids weight.



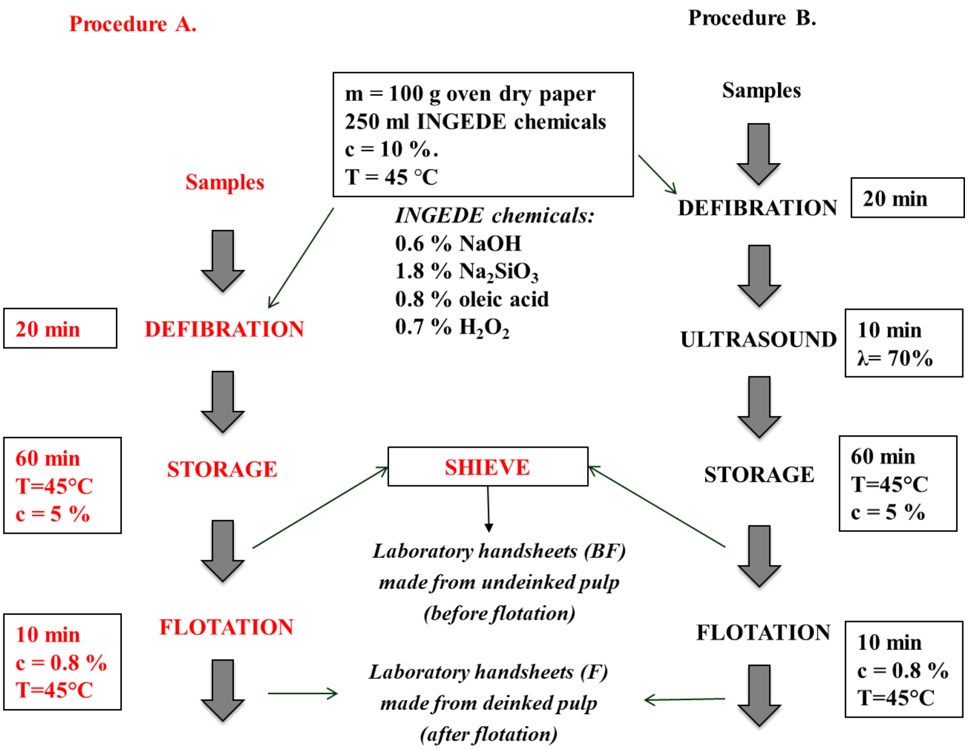


**Fig. 2: Processflow of the label backed with paper recycling by the INGEDE 11 method (procedure A) and INGEDE 11 method in combination with ultrasound (procedure B)**

## 2.3. Determination of optical properties

The efficiency of deinking flotation was evaluated by measuring optical properties of produced handsheets according to standard methods: ISO Brightness (ISO 2470), colorimetric properties (ISO 11475) and ISO Opacity (ISO 2471) on all manufactured laboratory sheets using Technydine Colour Touch 2 Spectrophotometer.

## 3. RESULTS AND DISSCUSSION

### 3.1. Sample preparation

During recycling process of polymer labels, several facts should be stated. First, during defibering of polymer labels on the paper support, the polymer labels were disintegrated into very large pieces. The print was printed on polymer and no ink particles was bonded to the fibres. It is importan to emphasize, that flotation was performed to remove large polymer pieces, not only for removal of ink particles. We assume that no ink particles were released into to pulp suspension during disintegration step. The main problem with recycling of paper with polymer label is a very difficult disintegration of present polymer. Additionally, the labels contain adhesives, causing another the problem with formation of sticky particles or stickies.

After disintegration step, from undeinked pulp, a certain amount of pulp suspension was taken and used for production of recycled paper. Due to the influence of adhesives and the polymer content in the sample, the pulp contained sticky particles that caused problems during handsheets drying at 92°C, i.e. during drying process paper was sticked to carrier board and cover sheet, a necessary parts for laboratory handsheets preparation. After drying process and after removal of carrier board and cover sheet, the paper was destroyed (Figure 3). Therefore, in order to obtain paper with satisfying quality, the sticky polymer particles had to be removed using a shieve.

**Fig. 3: The presentation of destroyed laboratory handsheet after drying process**

Furthermore, the pulp suspension was taken and used in flotation as it is an essential step in paper recycling process. Figure 4 shows the flotation process of paper with black and yellow labels. It can be seen that particles of the prints on the polymer surface are separated from the pulp regardless of their large surface area. This can be attributed to the hydrophobicity of the polymer used for printing. The particles were separated on the surface of the suspension together with other impurities, in the form of flotation foam.


**Fig. 4: Flotation process of paper with a) yellow and b) black labels**

Table 1 shows the weight of polymer that was removed from the undeinked pulp by a shieve, before the paper sheet making process, i.e. formation and drying.

**Tab. 1: The weight of removed printed polymer**

| Sample No. | INGEDE 11 | | INGEDE 11 + Ultrasound | |
|---|---|---|---|---|
| | m (polymer) | | | |
| | Yellow labels | Black labels | Yellow labels | Black labels |
| **1.** | 0.9 g | 1.5 g | 0.4 g | 0.6 g |
| **2.** | 0.8 g | 0.7 g | 2.1 g | 1.3 g |
| **3.** | 0.6 g | 0.3 g | 0.5 g | 0.7 g |



To produce 45 g/m$^2$ laboratory paper sheet (handsheet), certain quantities of printed polymer, as shown in Table 1., were separeated by a shieve and taken out from the pulp suspension. The values vary depending on the size of the separated printed polymer particles and it is evident that there is no repetitive sequence. Further research was carried out on the laboratory paper handsheets obtained by sieving the suspension and removing sticky polymer particles (for all samples made from undeinked pulp, before flotation process).

### 3.2. Somerville recyclability test

The Somerville recyclability test is a method of removing sticky particles. Table 2 shows the process efficiency values of the recycling black and yellow labels by the Somerville test. The results show that in both cases the waste (*reject*) is the same, about 55 - 57%. In both cases, the wastes are pieces of printed polymer. It is also evident that sample loss occurs during the process, probably the loss of fillers (10%). The fibers passed through a sieve, giving a clean suspension (without printed polymer) which is then dried and presented in this case as "*accept*".

**Tab. 2: Somerville test process efficiency**

| Somerville test | Sample | |
|---|---|---|
| | Yellow labels | Black labels |
| **$m_u$** | 18 g | 18 g |
| ***Reject*** | 10 g | 10.4 g |
| ***Accept*** | 6.3 g | 6.8 g |
| **Process efficiency , %** | 35 | 37 |

Where:

*Reject* - waste polymer (sieve residue)

*Accept* - cellulose fibers that have passed through a shieve

## 3.3. Determination of laboratory sheets optical properties

Figures 5 – 9 show the measured values of the optical properties for all recycled samples (made from pulp containing yellow and black labels). The results show the values of ISO brightness, colorimetric characteristics CIELAB *L**, *a**, *b** and ISO opacity, on samples made from undeinked (before flotation) and deinked pulp (after flotation) obtained by the INGEDE 11 recycling method and modified INGEDE 11method in combination with ultrasound. The results are the mean value of 10 measurements.

The obtained values of ISO brightness (Figure 5) show that both methods are equally effective in recycling. The brightness of all samples after flotation is about 81%, a slightly lower value is in the case of recycling black labels with the modified INGEDE method (in combination with ultrasound). By combining the INGEDE 11 method and ultrasound, in both cases, for black and yellow labels, the ISO brightness is lower than in the case when only the INGEDE 11 method was used.



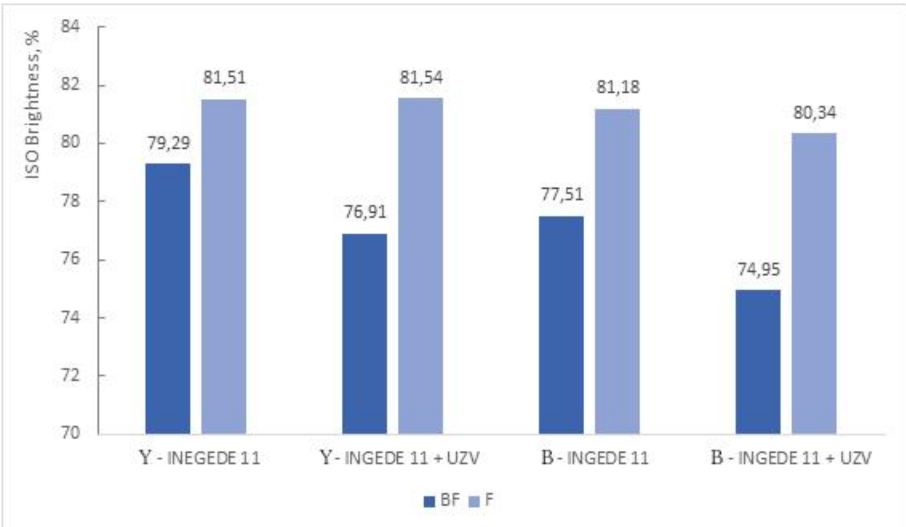


**Fig.5: ISO Brightness of samples obtained before and after flotation ((BF – undeinked pulp before flotation; F – deinked pulp after flotation; Y - Yellow labels; B – black labels)**

The increase of ISO brightness is presented in Table 3. The brightness increase was calculated using the Equation (1):

$$\text{Brightness increase, \%} = \text{ISO } B_F - \text{ISO } B_{BF} \quad (1)$$

where:

ISO $B_F$ – the value of brightness after flotation process,

ISO $B_{BF}$ – the value of brightness before flotation process.

**Tab. 3: The increase of brightness**

| | | Method | |
|---|---|---|---|
| | | **INGEDE 11** | **INGEDE 11 + ULATRASOUND** |
| **Brightness increase, %** | **Yellow labels** | 2.22 | 4.63 |
| | **Black labels** | 3.67 | 5.39 |

Figures 6 – 8 show the measured values of the colorimetric parameters *L**, *a** and *b**. The results (Figure 6) show that there was an increase in *L** value in all samples made from deinked pulp regardless to the recycling method (1-3% respectivley). A greater increase of 3% is seen in samples made by the INGEDE 11 method in combination with ultrasound. The values of the parameter *a** presented in Figure 7, show a decrease in the value in samples obtained from deinked pulp (50-69%). Based on the obtained results, we can see that the values of the laboratory paper handsheets of yellow labels obtined from undeinked pulp, regardless of the method of production, are much higher than the values of the samples of black labels for about 50%. Figure 8 show the values of the parameter *b**. The results show a decrease in values in the deinked samples made from yellow labels by the INGEDE 11 method. In other samles an increase in *b** values can be noticed. Comparing the used recycling methods, the increase of *b** values of samples is much higher with the use of ultrasound method.



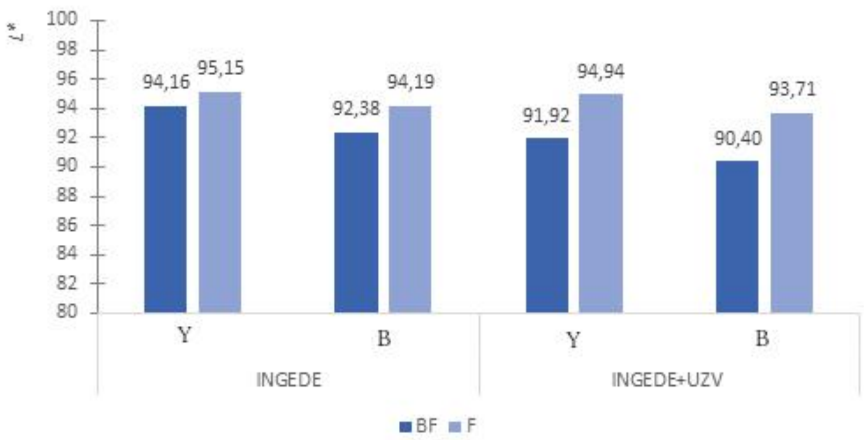


**Fig. 6: The obtained values of colorimetric parameter *L** (BF – undeinked pulp before flotation; F – deinked pulp after flotation; Y - Yellow labels; B – black labels)**

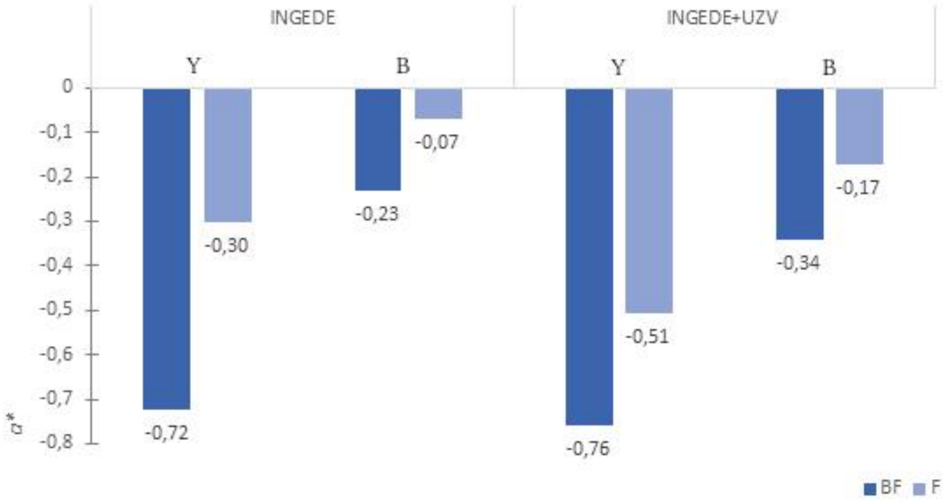


**Fig. 7: The obtained values of colorimetric parameter *a** (BF – undeinked pulp before flotation; F – deinked pulp after flotation; Y - Yellow labels; B – black labels)**

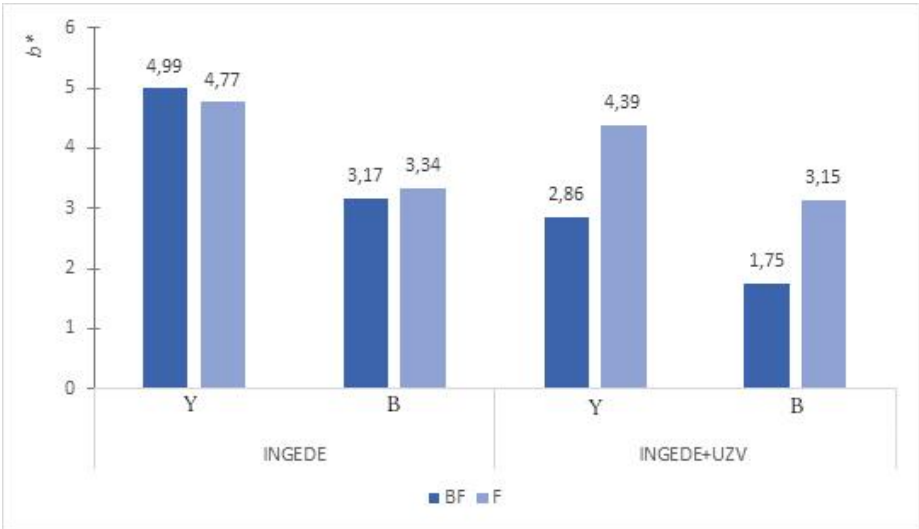


**Fig. 8: The obtained values of colorimetric parameter *b** (BF – undeinked pulp before flotation; F – deinked pulp after flotation; Y - Yellow labels; B – black labels)**

The results (Figure 9) show that after flotation the opacity of all samples decreases because there is a loss of filler during flotation process. The largest reduction was observed in the recycling of the black label sample by a combination of the INGEDE 11 method and ultrasound. The high loss of fillers durig flotation frocess can be attributed to sticky potential of labels (adhesive).

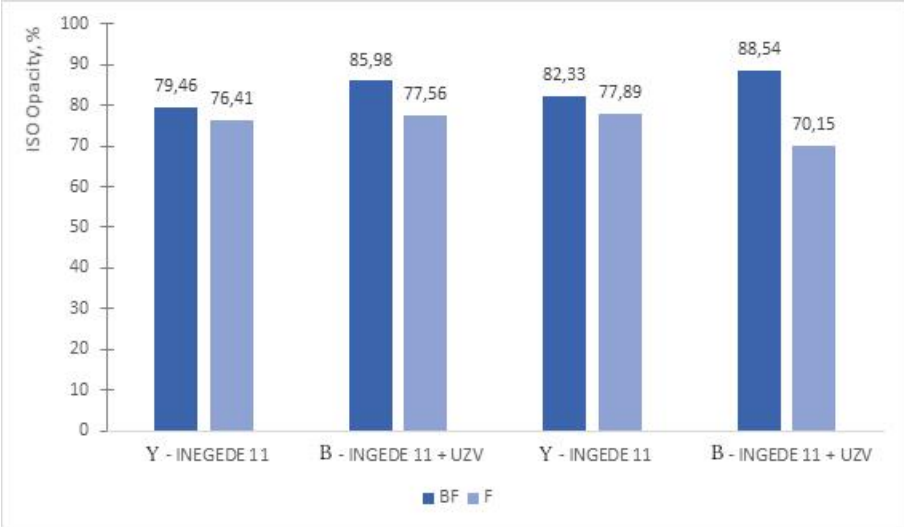


**Fig. 9: The obtained values of samples ISO Opacity (BF – undeinked pulp before flotation; F – deinked pulp after flotation; Y - Yellow labels; B – black labels)**

## 3.4. Determination of flotation yield

The results (Table 4) show that the flotation yield is almost the same in all samples (from 43-46%). The loss of the sample after flotation is significant, but we cannot talk about the loss of fibers, because almost all the printed polymer was removed by flotation. Therefore, such a high flotation yield is mainly due to the removal of the printed polymer not cellulose fibres.



**Tab. 4: Flotation yield**

| Recycling Method | INGEDE 11 | | INGEDE 11 + Ultrazvuk | |
|---|---|---|---|---|
| Sample | Yellow labels | Black labels | Yellow labels | Black labels |
| Flotation yield, % | 43.56 | 45.98 | 45.83 | 44.5 |

## 4. CONCUSION

This paper describes the possibility of recycling paper with self adhesive labes. From the presented results, and from the conducted practice of the process of laboratory sheets preparation, we can conclude that recycling of paper with self adhesive labes is possible but only because the prints on the polymer substrate show a hydrophobic nature, regardless of the fact that during disintegration they do not break into smaller particles but remain large. Although the prints together with the polymer substrate are separated during deinking flotation in the form of flotation froth (foam), to make laboratory handsheets from undeinked pulp, it is necessary to first remove printed polymer by a shieve. The reason for this is that present adhesives, which during disintegration are not removed or dissolved during the recycling process, cause certain problems. The problems are related to formation and drying of paper sheets ad formation od sticky particles in the pulp. The resulting paper is destroyed after drying, so before the process of forming a sheet of paper, the labels need to be separated by some physical means such as sieving. Based on the obtained results of process efficiency, it is evident that the measured values obtained by all methods are the same, which means that the addition of chemicals in the deinking process does not affect the process efficiency, nor ultrasonic pretreatment that did not improve the label recycling process. Self adhesive labels made from polymers and applied on papers are very difficult to recycled with classic paper recycling methods, since the used polymer does not disintegrate in the paper recycling process and adhesives form sticky particles. However, with the increase in the consumption of this kind of labels, there is a possibility of their excessive accumulation as a waste material in the stream of paper for recycling, which in the end may influence paper recycling process. From another point of view, the fibers in paper support are valuable raw material in papermaking. Therefore, it is necessary to investigate the recycling technologies, challenges, and possibilities of polymer self adhesive labels sticked to papers. Although there is a large loss of input material during recycling, and it mainly refers to the polymer, recycling of paper with labels is possible because the paper on which the labels were attached is separated and this paper is a potential new raw material for fiber production. Additionally, the choice of proper recycling method can affect the properties of produced recycled pulp.

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