Material Recovering from Offset Prints on Paper with Alternative Fibers

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Abstract: The development of non-wood fibers has been one of strategies to ensure a sustainable fiber supply, including plantation management, reforestation and recycling. Some of alternative fibers are algae that can be used as raw material for papermaking. The Shiro Alga Carta papers made from algae in combination with Forest Stewardship Council certified fibers (FSC - fibers) were used in this research. Offset inks used in printing are vegetable-based (different amount of renewable raw materials) and mineral oil-based.

The aim of this research was to contribute to the explanation of the ink/substrate interaction in printing on the detachment of ink from substrate and removal, as well as to study the recovered fiber characteristics. In our research we used the three-loops method.

Generally, satisfactory characteristics of the recovered fibers were obtained in experimental conditions for all samples, as it is expected from environmentally friendly, with algae speckled paper. The research was supported by setting environmental sustainability.

Keywords: Paper, algae, different inks, re-pulping, fibers recovering, handsheets characteristic

Introduction

The growing demand for sustainable business is significant in the graphic industry. The waste and influence on environment depends on the printing technics and the used materials. In offset printing techniques waste includes: substrates, inks, fountain solution, metal plates, volatile organic compounds, biocides, varnish and energy (Bolanča Mirković, Bolanča, 2007; Bolanča Mirković, et al. 2011; Amon-Train, et al., 2012; Bolanča Mirković, et al., 2015; Bolanča, Bolanča Mirković, 2017).

Some life cycle reports on offset printed matter points to the paper production stage, with forestry and pulp production and delivering the paper product, as the dominating contributor to the estimated potential environmental impacts from the LCA (life cycle assessment) of offset prints (IFRAS, 1998; Larsen et al., 2006; Bousquin et al., 2011). The analysis of the impacts on damage level shows that the paper production is significant to the AoPs (damage areas of protection): global warming, acidification and eutrophication, and dominates the specific contribution to the AoP ecosystem. The printing stage has a higher share both to human health and resources related impact categories.

Another source states that in the life cycle assessment it is found that the offset printing process is the largest contributor to the environmental impact, more than the impact of paper and inks (printing 52%, paper31%, ink 17%) (Dougherty, 2008).

Larsen and coauthors studied the life cycle assessment of offset printed matter with EDIP 97 method. Emphasis in that research was on how important are emissions of chemicals (Larson et al., 2009)? This study includes the newest knowledge about emissions from the printing industry and knowledge about the composition of the used printing materials. The results show that the use of paper no longer becomes the overall dominating factor causing the environmental impact.

The pulp and paper industry is the resource intensive industry and contributes to many environmental problems, including global warming, ecotoxicity, human toxicity, photochemical oxidation, acidification, eutophication and solid waste (Bajpai, 2010). Ecological advantage is focused on pollution prevention rather than on end-of-pipe clean up. Sustainable development includes cleaner production measures in raw material storage and preparation, in pulping processes, in bleaching, recovery, papermaking, emission and in recycling.

KrishnaManda and Patel investigated whether the use of micro and nano TiO_2 as coating and different pulp types could bring savings in wood, energy, GHG emissions in comparison with conventional printing paper (KrishnaManda, Patel, 2012). The results show that the nanoparticle coated recovered fiber paper saves nonrenewable energy use by 100% and generates GHG emission reduction by 75%.

By the application of innovative manufacturing strategies using advanced sheet structure design and fiber modifications, the reduction of GHG emission by 22.9 % for SC paper and by 20.3 % for LWC paper can be achieved. (Jorgel, Juan, 2015).

Recycled paper is better for the environment than virgin paper: it preserves forests, conserves resources, generates less pollution during manufacturing and reduces solid waste. In literature, this issue is being investigated a lot, and Baypan states that for a tonne of recycled paper, compared to a tonne of virgin paper, it reduces the use of: wood 100%, wastewater 33%, energy consumption 27%, air particulate emissions 28% and solid waste 54% (Bajpai, 2014).

Recycling of waste printed matter has also negative environmental impacts: it requires the removal of inks from prints and plastic polymer from office waste paper and often contains dioxins (Kesalkar, 2012). Furthermore, wastepaper recycling produces sludge that contains small fibers, ink from de-inking process and fillers. Printing inks contain heavy metals (copper, lead, zinc, chromium and cadmium) and solvents.

Michaud and co-authors studied the carbon balances between paper recycling, the disposal of landfill and incineration (Michaud et al. 2010). They concluded and published within the framework Waste and Resources Action Program (WRAP) that recycling 1 tonne of paper will avoid 1.4 tonnes of CO_2 equivalent compared with landfill, and 0,62 tonnes of CO_2 equivalent compared with incineration.

In recent years with growing of environmental awareness, there is an increasing number of investigations in the field of alternative non-wood fibers for papermaking. In literature it has been reported on wheat straw, sisal, banana sten, kenaf, corn, bamboo, elephant grass as the basis of pulp and paper (Shivhare et al., 2012; Sibani et al., 2012, Qin et al., 2011; Mosseto et al, 2010; Egbewole, et al., 2015; Gomes, et al,. 2013). LCA studies show that this type of fiber little contributes to almost none of the environmental impact categories compared with pulp mill operations. The energy and environmental impacts of both pulping and paper can dominate the overall environmental impacts; these impacts can matter more than the choice of fiber and should be considered in an overall evaluation of agricultural inputs (Favero et al., 2017). New, alternative pulping processes were studied because of highly contaminant classical treatments for obtained paper with acceptable properties coming from unconventional pulping raw materials treated with alternative processes which is a way to sustainable and economic production.

Algae are an alternative aquatic raw material for papermaking, and it is a solution to global environmental issues: deforestation and global warming. Algae contain low lignin-like compounds in cell walls, and there are no problems associated with lignin removal as with cellulose obtained from trees and other vascular plants (Knoshaug, et al. 2013).

Paper produced from red algae as Gelidium takes lower cooking temperature, shorter time and less chemical usage compared to wood pulp (Mukherjee, Prakash Keshri, 2018). Algae fibers are finer and more uniform in length, which improves its utilization as reinforcing fibers in the manufacturing of both virgin and recycled paper. Lee et al. found that the bleached red algae fiber showed higher thermal stability than that of crystalline cellulose (Lee et al., 2008).

Seo and coauthors compared the algae-based handsheets to wood handsheet of the same weight (Seo, et al., 2010). They found that density and breaking length were lower for the algal handsheets, while the brightness and stretch values were comparable, and smoothness and freeness were higher, but you can mix the fibers with softwood fibers for better paper properties.

A particular interest is devoted to the study of green algae and seaweeds, respectively (*Cladophora sp., Ulva sp., Rhizoclonium)* as a raw material for paper making. Thus, pulping procedure, physicochemical properties, structural characteristics, unit cell parameters, degree of crystallinity and mechanical properties of algae cellulose were investigated (Lopez et al. 2013; Martone et al., 2009; Mukherjee, Prakash Keshri, 2018, Marsin, 2005; Mihranyan, 2010).

In our extensive research we used a printing substrate containing a certain amount of green algae genus Ulva. These algae are found in the Adriatic Sea because of eutrophication. In this article, we are presenting only a part of our results relating to the domain of the offset print life cycle. The aim of this research was to make a contribution to the explanation of the ink/substrate interaction in printing on the detachment of ink from the substrate and the removal from pulp, as well as to study the recovered fibers characteristic. The research was supported by settings of the environmental sustainability.

Materials and methods

Samples were made on the five-color offset machine with a coating unit Roland 705. The printing form contained different printing elements: a standard CMYK step wedge in the 10-100% tone value range, a standard ISO illustration for visual control, textual positive and negative microelements, wedges to determine greyness and the standard wedge with 378 patches to produce ICC profiles and 3D gamut.

In the research materials based on renewable raw materials in line with one of the essential sustainable development settings were used. The Shiro Alga Carta paper made from algal blooms which grow in the Adriatic Sea (patented and manufactured by Favini, Italy) was used in this research (marked P_2) FAVINI, 2012). Eutrophication is a process which causes an excessive algal growth and it becomes enriched with an increase in pollution, organic effluents and temperature. Algae were dried and ground in a colloid mill to a size less than 500µm.The algae were used in partial substitution of pulp and combined with FSC fibers (FSC - Forest Stewardship Council, certified forest, so they are grown according to sustainability principles). This paper is environmentally friendly grayish-green speckled paper where the speckles are the milled algae.

The prints were prepared with the offset inks different composition, produced by SunChemical® Europe. Inks that were used are available as a four-process color offset ink set. The inks marked withB_1 contain 78-82% of renewable raw materials. These inks are based on an innovative resin/oil combination. Inks dry by absorption and oxidation. Inks marked with B_2 are vegetable-based and free of mineral oils and dry by penetration and to a high degree by oxidation. The inks marked with B_3 are mineral oil-based and are free of cobalt based drying catalysts. These inks are dried by penetration and to a high degree by oxidation.

Print varnishing was performed with water-based dispersion varnish Hi-Tech. Coat W6000 Heidelberg Group (marked L_2). According to the manufacturer's statement, the amount of lead, chromium, cadmium and mercury is in accordance with the total maximum below threshold given by Directive 94/62/EC. The regulation concerning the restriction of use of certain epoxy derivatives was applied.

The flow of the experiment is shown in Fig. 1. The fibrous material recovery process in one part uses INGEDE method 11 (INGEDE 11, 2012). In our research we used re-pulping (loop I, loop II, loop III), rather than collector oleic acid and flotation method, like that described in INGEDE 11 method.

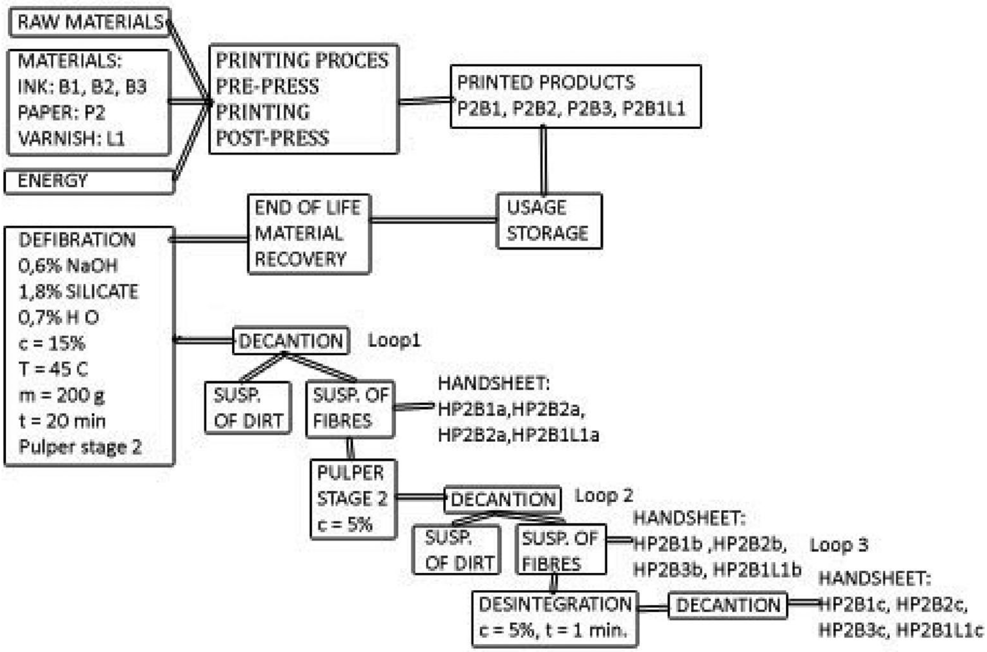

Fig. 1 – Scheme of the experimental flow

The handsheets were made by using the Rapid-Köthen sheet former according to standard ISO 5269-2 (ISO 5269-2, 2002) The following methods were used for measuring the optical characteristics of laboratory handsheets: diffuse blue reflectance factor according to ISO 2470 and effective residual ink concentration-ERIC according to TAPPI T 567 pm-97 (ISO 2470, 2005, TAPPI T 567, 2009).

The count of the residual dirt particles and area were assessed by using the Spec*Scan Apogee System image analysis software (ISO 13322, 2014). This system utilizes a scanner to digitalize an image. The threshold value (100), white level (75) and black level (65) were chosen after comparing the computer images to the handsheets.

Environmental sustainability is significant for development, and it includes energy and material flow, closed loop systems and clean technology. Having that in mind, and in order to optimize the reusing fibers from substrate which contain algae, research was conducted to determine the characteristics of the obtained fibers. Thus, print P_2B_1 (substrate with algae, ink containing about 80% of the renewable raw material) is overprinted with water dispersion varnish. The aim was to determine the influence of varnish on the efficiency of the material return and the characteristics of the obtained fibers. The specks count on handsheets from loop I, II and III according to the scheme of the experimental flow is shown in Fig. 2.

The results showed a noticeable dependence of the process stages on the total count of specks on handsheets as follows: $HP_2\,B_{1a}/HP_2\,B_{1b}$ = 39.8%, $HP_2\,B_{1b}/HP_2\,B_{1c}$ =

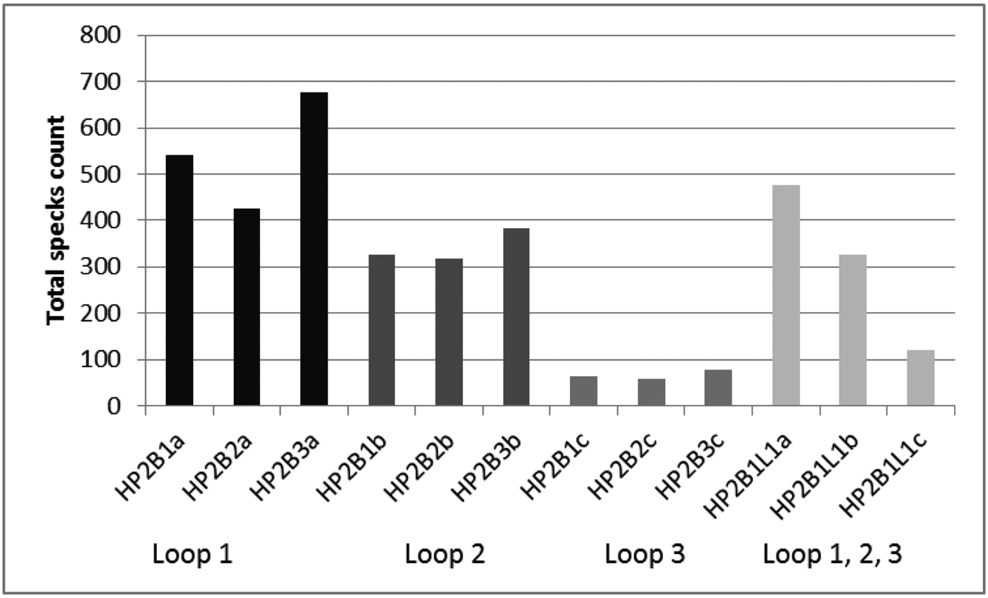

Fig. 2 – Total specks count on handsheet versus process stages

88.4%; HP_2B_{2a}/HP_2B_{2b} = 19.3%, HP_2B_{2b}/HP_2B_{2c} = 86.6%; HP_2B_{3a}/HP_2B_{3b} = 43.6%, HP_2B_{3b}/HP_2B_{3c} = 88.5%.The efficiency of the specks removal from fibers is dependent on the number of loops and on the formulation of inks. Thus, the efficiency of the specks removal is increased in the third loop in comparison to the second loop as follows: HP_2B_1=48.6%, HP_2B_2=64.2%, HP_2B_3=44.9%. Specks removal efficiency is achieved by the described process including all stages, as follows: HP_2B_1= 88.4%, HP_2B_2= 86.1%, HP_2B_3= 88.5%.

The highest fragmentation of inks was noted for prints with mineral oil-based inks ($B_{3\ total\ specks}$ = 677), and the lowest for prints with the vegetable-basis inks without mineral oil ($B_{2\ total\ specks}$ = 426).

The print with ink containing about 80% of the renewable raw material is overprinted with water-based dispersion varnish. Varnishing is used to heighten a gloss or matt finish and to protect the surface of a printed product from scuffs, scratches and fingerprints. Varnishing affects the total count of specks in all stages of the process. The efficiency of the specks removal is smaller for the varnished pattern compared to non-varnished as follows: loop1/loop2 = -8.4 %, loop 2/loop 3 = -15.2%.

The purpose of understanding the mechanism of the ink detachment from the substrate is determined. The specks surface that they occupy the handsheets made of fiber from different stages of the process is described (Fig. 3).

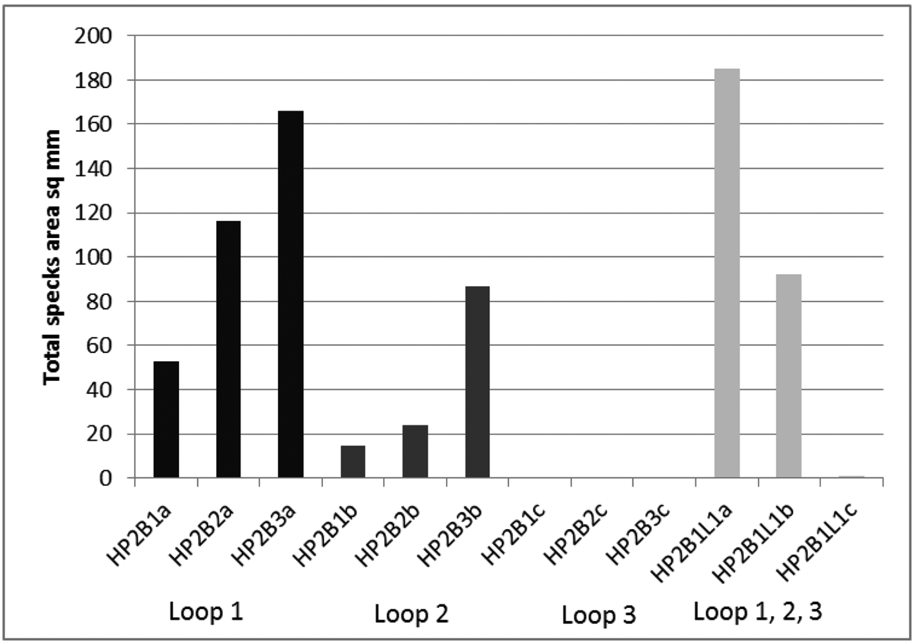

Fig. 3 – Total specks area on handsheet versus process stages

These results confirmed the previously displayed dependence of the loop number as well as inks formulations on the detachment and removal of the inks from substrate. The largest total area covered with specks was determined for the handsheet made from recovered fibers from sample $P_2 B_1 L_1$ (substrate with algae, ink containing about 80% of the removable raw material, prints overprinted with water-based dispersion varnish) after loop 1 ($HP_2 B_1 L_{1\ a}$ = 185.1 mm^2).

The handsheet made from recovered fibers from the non-varnish sample $P_2 B_{3a}$ (substrate with algae, mineral oil-based ink) after loop1 ($HP_2 B_{3a}$ = 166.00mm^2) follows.

The dependence of the total area of specks on the number of the loop and the formulation of inks is as follows $HP_2 B_{1a}/HP_2 B_{1b}$ = 77.3%, $HP_2 B_{1b}/HP_2 B_{1c}$ = 95.6%; $HP_2 B_{2a}/HP_2 B_{2b}$ = 79.7%, $HP_2B_{2b}/HP_2 B_{2c}$ = 97.9%; $HP_2B_{3a}/HP_2 B_{3b}$ = 4.9%, $HP_2B_{3b}/HP_2 B_{3c}$ = 99.3%.

The efficiency of the specks removal is increased in the third loop in comparison to the second loop as follows: $HP_2B_{1\ =}$= 9.2%, HP_2B_2= 18.6%, HP_2B_3 = 42.2%. In summary, the specks removal efficiency is achieved by the described process including all stages is as follows: HP_2B_1 = 95.6% HP_2B_2 = 97.9% and HP_2B_3 = 99.3%.

For the procedure of recovering fiber one of the primary quality criteria is cleanliness of the handsheet. The criteria for cleanliness are a minimum dirt count or specks. The optical inhomogeneity of the handsheet is significant for the specks of a size >= 0.04mm^2. The specks of a size < 0.04mm^2 are primarily relative to the optical properties of the handseet and refer to the surface grayness. Figures 4 and 5 show ink removable efficiency for the described phases of the fibers recovery process, for the specks class of a size >= 0.04mm^2 and < 0.04mm^2.

There is a significant difference in the efficiency of the process with regard to the size of the specks. Generally, specks in the class >= 0.04mm^2 are better removed, compared to specks in the class size < 0.04mm^2 by the values as follows: $HP_2B_{1ac,>= 0.04mm2\ -\ <\ 0.04mm2}$=9.5%, $HP_2B_{2ac,>= 0.04mm2\ -\ <\ 0.04mm2}$=12.6% and $HP_2B_{3ac,>= 0.04mm2\ -\ <\ 0.04mm2}$=7.0%. The larger specks compared to the smaller ones can be removed easily, considering that the process is mechanical precipitation.

The ERIC (effective residual ink concentration) measurement is dependent on the distribution of ink particle sizes, being most effective for submicron particles. Determination of the effective residual ink concentration on a handsheet requires the measurements of reflectance in the infrared area of the spectrum where the absorption coefficient for the ink is several orders of magnitude greater than the absorption coefficient for the fiber, filler, fines and other components. The largest difference in the ERIC appears system $HP_2B_{2a} – HP_2B_{2c}$ = 317.2.

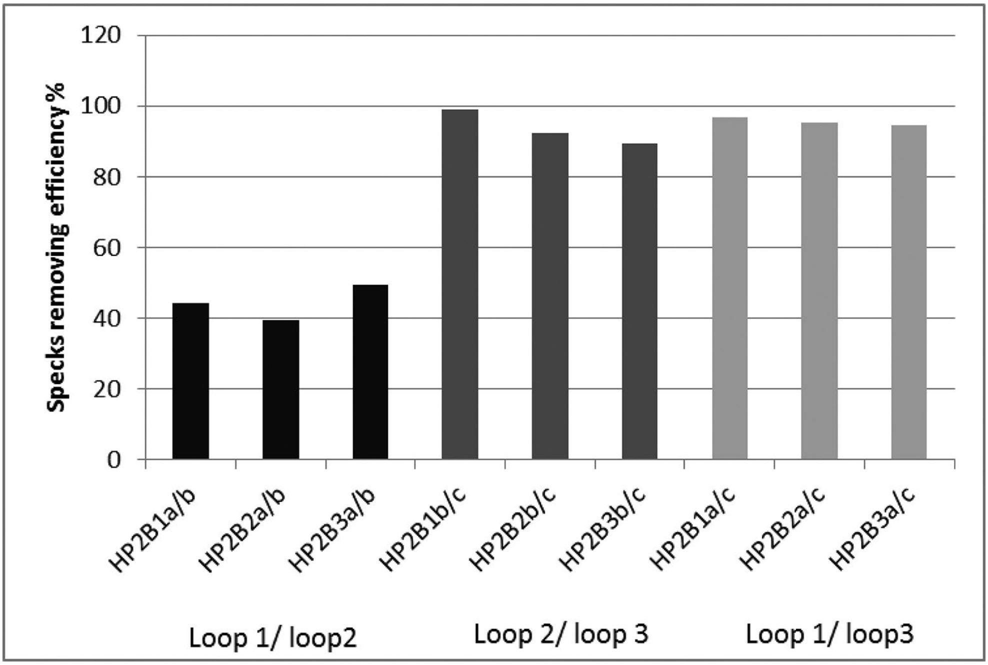

Fig. 4 – Specks removing efficiency for the class of a size >= 0.04mm^2

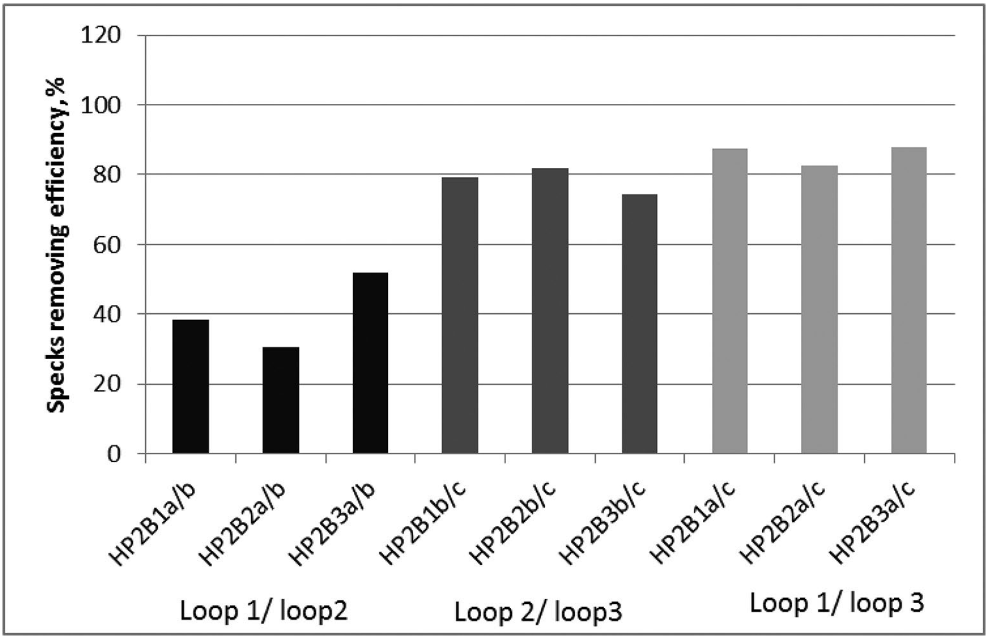

Fig. 5 – Specks removing efficiency for the class of a size < 0.04mm^2

The same sample confirms the highest gain in brightness ($B_{gain\ HP2B1} = 10.3$). The differences in the ERIC successfully show the efficiency of the described stages in the experimental flow, and confirm the results obtained using other methods.

The offset printing process and drying mechanisms influence on ink detachment from substrate. Sheet fed offset ink has oxidable components in the formulation because it is necessary for the printing process. Vegetable-based inks, especially some unsaturated vegetable oils, can cause ink detachment problems and specks. Some resins added into the ink can also lead to the strong attachment of the ink onto fibers.

The organic structure of mineral-oil based inks was relatively unchanged due to oxidation compared with rapid oxidation and cross-linking of vegetable- based inks. For mineral oil-based inks the drying process is absorption with lingering oxidation.

Conclusions

Based on the results obtained from the three-loop process, recovering fibers from the offset prints on substrates made from FSC fibers and algal blooms in combination with different ink compositions, the following conclusions were reached.

Just a little better efficiency of specks removing with the three-loop process was determined for prints with mineral-oil based ink on the substrate with algae in comparison with the other two samples. A slightly lower efficiency of specks removing was determined for prints with vegetable - based ink because the oxidable components by drying the ink in printing causes the cross- linking structure to appear. Improved vegetable ink appears almost as good results (better for speck contamination) compared with mineral oil–based ink.

It was found that specks in the class >= $0.04mm^2$ are better removed compared to specks in the class size < $0.04mm^2$, especially in the print with ink containing more than 70% of the renewable raw material. Handsheets obtained from the recovered fibers of this print have the highest Δ ERIC and brightness gains.

The print with ink containing about 80% of the renewable raw material overprinted with water-based dispersion varnish has a slightly lower efficiency of speck removing compared to the unvarnished prints.

Generally, satisfactory characteristics of the recovered fiber were obtained in experimental conditions for all samples, as it is expected from environmentally friendly, with algae speckled paper.

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