

Ecologically Sustainable Printing: Aspects of Printing Materials

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Abstract: 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 for this research. Three offset inks were used in printing: vegetable-based inks with different amount of renewable raw materials and mineral oil - based ink. Three series of prints were made: non-varnished, varnished with water-based dispersion, and varnished with UV cured varnish. The possibility of reusing fibers was determined by methods of re-pulping (loop I, loop II, loop III). The vegetable based ink toward improved structure give better results in terms of brightness, ink content and dirt count and area compared with the mineral oil-based ink. Something lower brightness gain and ΔERIC is obtained with water-based dispersion overprint samples. The lowest results are obtained for samples overprinted with UV cured varnish. The results of the research contribute to the explanation of paper/ink/varnish interaction on the reuse of the fibers, which takes into the area of the ecological sustainable development.

Keywords: Algae; cardboard; ink; offset printing, recovered fibers; re-pulping; varnish

1 INTRODUCTION

Sustainability and environment issues are one of the most important topics for strategic business and product development decisions. The environmental sustainability refers conditions where neither on a planetary nor on a regional level human activity, disturb the natural cycles more than planetary resilience allows [1]. Sustainability for development includes flows of energy and materials, clean technology, closed loop systems, quality and economic and social [2].

The environment impacts of the printing industry are significant and is characterised by the use of energy and environmental pollution due to chemical intensive processes. In the recent past, a lot of research papers have addressed sustainability issues of printing. Within each printing technic, a variety of chemicals are used, depending on the types of operation involved.

Offset printing is a chemical intensive technic that produces many types of waste [3]. The main environmental issue of the offset printing are: use of non-renewable resource (mineral pigments in paper, mineral oils inks and

solvents, metal plates, metal and plastic in equipment), use toxic or harmful substances (additives in inks and adhesives, biocides in fountain solution), VOC (volatile organic compounds) emissions (from inks mixing, and drying, evaporation from fountain solution, evaporation from cleaning solutions, blanket washes), toxic waste (ink waste, cleaning solvent waste), regenerative waste (paper, unacceptable prints), use of energy (production equipment, ink drying, transport) and transport emissions (supply chain of paper, ink, varnish, delivery of printed matter, transport of waste) [4-8].

Life cycle assessment (LCA) studies the environmental aspects and potential impact of a product through its life from raw material exploitation through production, use and disposal (Fig.1).

Pre-press processes today are digital, and now not used photographic processes. Platemaking utilizes computer to plate (CTP) techniques, where the printing form is exposed directly from digital data with laser or LED arrays [9]. The further procedure depends on whether it will be used, plate processing chemicals, which must be treated as hazardous waste.

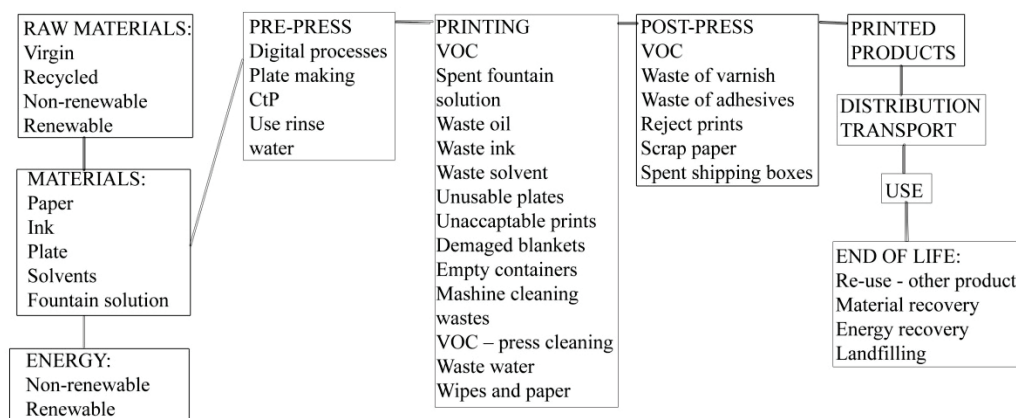


Figure 1 LCA of the offset printing and types of waste

The printing process has environmental concerns from the use of energy and material resources. The life cycle assessment found that the sheet fed offset printing process was the largest contributor to the environmental impact of most printed materials (printing 52%, paper 31%, ink

17%), and the dominating impact expressed in acute ecotoxicity [10]. Emission consists of fugitive VOC emission, and consumption of VOC is 3 kg/tonne product [3]. VOC containing chemicals can in many cases be substituted with other agents that have lower

environmentally and health effects [11]. UV inks are designated to suitable drying with LED-UV lighting and reduce electricity consumption, and can contribute to a reduction in VOC.

In term of the resource usage, paper is still dominant at 48% [12]. The main issues are the impact of forestry to biodiversity, the energy consumption in paper making and the emissions to air and water from chemical pulping, deinking and bleaching.

Due to the current environmental settings arise an attention is being led to the development of non-wood fibers. Alternative fibers have been one of strategies to a sustainable fiber supply including recycling, reforestation programs and plantation management [13, 14]. Some of the new materials are algae that can be used as the raw material for papermaking.

The lignin content in the algae cell wall is so low that there are not problems associated with lignin removal as in the conventional extraction of cellulose in papermaking [14, 15]. Lopez and coauthors estimated the amount of lignin and holocellulose in dried algae pulp (bloom algae *Ulva sp.* and *Cladophoraspp.*) [16]. Results showed that the algae have low lignin-like compounds and solvent-soluble substances content which supposes advantage over the current cellulose extraction method.

Some Families *Gelidialian* red algae can also be used as raw materials in papermaking. Seo and co-authors compared the algae-based handsheets to wood pulp handsheets of the same weight [17]. The density and breaking length were lower for the algal handsheets, while the brightness and stretch values were comparable, and opacity, smoothness and freeness were higher.

The aim of this paper is to determine the impact of more environmentally friendly graphic materials (substrates, inks, varnishes) on the quality of recovered fibers suitable for obtaining fine graphic paper. Research uses paper made from FCN fibers and green algae genus *Ulva*. Detachment of ink from substrate and removal from the system is achieved using a three-loop method without the addition of surface active chemicals and flotation. These studies are a contribution to ecological sustainability in the field of offset printing.

2 EXPERIMENTAL

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, textual positive and negative microelements, and the standard wedge for the production of ICC profiles and 3D gamut.

The Shiro Alga Carta paper made from algal blooms which grown in the Adriatic Sea (patented and manufacturing by Favini, Italy) was used for this research (marked P₂) [18]. The specific interest in green algae is justified by unwanted global environmental problems associated with seasonal blooms. These algae compromising the ecosystem balance and they accumulated in the beach. On the other hand, the physicochemical properties and the use of this unique material are highly beneficial. One of algae applications is in papermaking, considering of their structural characteristics. The algae were collected from the lagoon, dried and ground in a colloid mill to a size less than 500 µm [18]. 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 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 B₁ contain 78-82% of renewable raw materials [19]. These inks are based on an innovative resin/oil combination. Inks dry by absorption and oxidation. Inks marked B₂ are vegetable-based, free of mineral oils, dry by penetration and to a high degree by oxidation [20]. The inks marked B₃ are mineral oil-based, and are free of cobalt based drying catalysts [21]. These inks are drying 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 L₁).

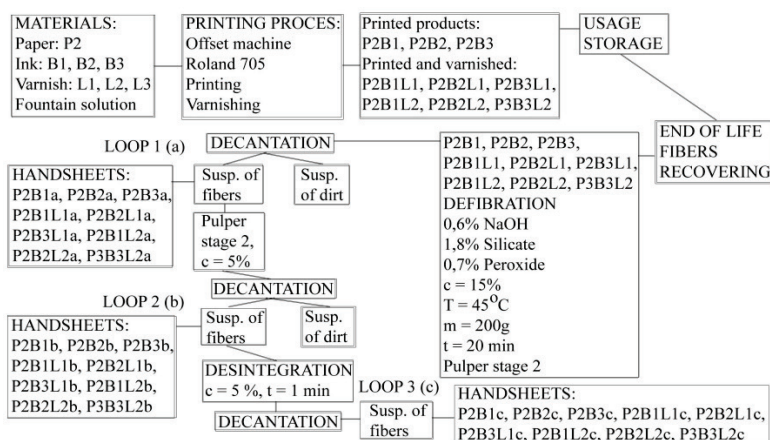


Figure 2 The flow of the experiment

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 [22]. The regulation concerning the restriction of use of certain epoxy derivatives has been applied.

In addition, the UV cured varnish VP 1038 high gloss, VergamGH (marked L2) was used. The characteristics of this varnish are: highly reactive, photopolymer able acrylate system, VOC free, reduced odor and optimal wetting properties [23]. The film building results from radical polymerization by high-pressure mercury lamp. When applying the UV cured varnish on prints produced by the mentioned offset inks, because of possible problems with ink drying and appearing the undesirable effect of gloss decrease (the drawback effect), the primer was used to solve the problem. After that treatment, the UV varnishing followed.

The fibrous material recovery process in one part uses INGEDE method 11 [24]. In our research we use re-pulping (loop I - marked a, loop II - marked b, loop III - marked c) rather than collector oleic acid and flotation method, like that is described in INGEDE 11 method. The flow of the experiment is shown in Fig. 2.

The handsheets were made by the using the Rapid-Köthen sheet former, according to the standard ISO 5269-2 [25]. The following methods were used for the measuring of the optical characteristics of laboratory handsheets: the diffuse blue reflectance factor according to ISO 2470 and effective residual ink concentration-ERIC according to TAPPI T 567 pm-97 [26, 27].

The count of the residual dirt particles and area were assessed by using the Spec*Scan Apogee System image analysis software (ISO 13322, 2014) [28]. 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.

3 RESULTS AND DISCUSSION

The aim was to determine the influence of varnish on characteristics of the recovered fibers. The quality parameter which characterizing the recovery pulp in cleanliness is dirt count and area. The total dirt area on handsheets obtained from loop I, II and III, for HP₂B₁, HP₂B₂, HP₂B₃, HP₂B₁L₁, HP₂B₂L₁ and HP₂B₃L₁ according to the scheme of the experimental flow, is shown in Fig. 3a.

The results showed noticeable dependence of the kind of inks on the total dirt area on handsheets from the first loop as follows: HP₂B₁ = 60.6 mm², HP₂B₂ = 36,0 mm² and HP₂B_{3a} = 87,7 mm². During the process the total dirt area is reduced: HP₂B_{1b}/HP₂B_{1a} = 51,3%; HP₂B_{2b}/HP₂B_{2a} = 59,4%; HP₂B_{3b}/HP₂B_{3a} = 83,9%; HP₂B_{1c}/HP₂B_{1a} = 98,7%; HP₂B_{2c}/HP₂B_{2a} = 98,1% and HP₂B_{3c}/HP₂B_{3a} = 98,8%. The loop II decreases the most total dirt area on the handshet HP₂B_{3b} compared to the handshets HP₂B_{1b} and HP₂B_{2b} while the loop III are achieved almost the same result for all studied inks.

Prints P₂B₁ and P₂B₃ overprinted with water-based dispersion varnish (L₁) are less efficient in the re-pulping process-loop II/loop I compared to non-varnished prints (HP₂B_{1b}/HP₂B_{1a} = 51,3% and HP₂B_{1L1b}/HP₂B_{1L1a} = 36,6%; HP₂B_{3b}/HP₂B_{3a} = 88,9% and HP₂B_{3L1b}/HP₂B_{3L1a} = 42,6%). Prints varnish with water-based dispersion varnish does not affect the efficiency of the re-pulping process when comparing the handsheets obtained from loop I and loop II.

The total dirt area on handsheets from loop I, II and III for prints P₂B₁L₁, P₂B₂L₁, P₂B₃L₁, P₂B₁L₂, P₂B₂L₂ and P₂B₃L₂ according to the scheme of the experimental flow, is shown in Fig. 3b. Dirt removal from fibers is slightly less in case of prints overprinted with UV cured varnish compared to water-based varnish as follows: HP₂B₂L_{2b}/HP₂B₂L_{2a} = 12,40%; HP₂B₂L_{1b}/HP₂B₂L_{1a} = 65,23%; HP₂B₃L_{2b}/HP₂B₃L_{2a} = 28,6%; HP₂B₃L_{1b}/HP₂B₃L_{1a} = 42,6%. Better results are received when comparing total dirt area on handsheets obtained from loop I and loop III: HP₂B₁L_{1c}/HP₂B₁L_{1a} = 9,37%; HP₂B₁L_{2c}/HP₂B₂L_{2a} = 95,57%; HP₂B₂L_{1c}/HP₂B₂L_{1a} = 98,64%; HP₂B₂L_{2c}/HP₂B₂L_{2a} = 93,37; HP₂B₃L_{1c}/HP₂B₃L_{1a} = 99,23%; HP₂B₃L_{2c}/HP₂B₃L_{2a} = 9,43%.

The total dirt count on handsheets HP₂B₁, HP₂B₂, HP₂B₃, HP₂B₁L₁, HP₂B₂L₁ and HP₂B₃L₁ obtained from loop I, II and III fibers, for prints P₂B₁, P₂B₂, P₂B₃, P₂B₁L₁, P₂B₂L₁ and P₂B₃L₁ according to the scheme of the experimental flow, is shown in Fig. 3c.

The results showed dependence of the kind of inks on the total dirt count on handsheets from the first loop as follows: HP₂B_{1a} = 433, HP₂B_{2a} = 233 and HP₂B_{3a} = 398. The efficiency of the specks removal is increased in the second and the third loop in comparison to the first loop as follows: HP₂B_{1a}/HP₂B_{1b} = 35,3%; HP₂B_{1b}/HP₂B_{1c} = 7,2%; HP₂B_{2a}/HP₂B_{2b} = 36,9%; HP₂B_{2b}/HP₂B_{2c} = 66,1%; HP₂B_{3a}/HP₂B_{3b} = 30,9%; HP₂B_{3b}/HP₂B_{3c} = 81,4%.

The efficiency of the dirt removal is mainly smaller for the water-based varnished pattern compared to non-varnished and the biggest deviation is in the case of B₃ ink HP₂B_{3c/a}/HP₂B_{3L1c/a} = -4,78%.

Quite different results are determined by comparing the handsheet made of fibers from treatment of prints overprinted with water-based dispersion varnish compared to those overprinted with primer and UV cured varnish: HP₂B₁L_{1a/b} - HP₂B₁L_{2a/b} = 29,77%, HP₂B₂L_{1a/b} - HP₂B₂L_{2a/b} = 6,01% and HP₂B₃L_{1a/b} - HP₂B₃L_{2a/b} = 8,49% (Fig. 3d).

Total count of dirt are increased on the handsheets HP₂B₁L_{2c}, HP₂B₂L_{2c} and HP₂B₃L_{2c} (loop III), which is not the case with all other researched samples. A detailed insight is obtained by tracking the count of dirt into 26 size classes and estimated equivalent black area of a gray or colored speck is smaller than its actual area in inverse proportion to the intensity of its color contrast with its background (Figs. 4a, 4b, 5a, 5b).

Varnishing is used to heighten a gloss or matt finish and to protect the surface of a printed product from scuffs, scratches and fingerprints. When deciding whether a coating should be used, it is important to consider the application of prints, its required durability and life span. Compared to aqueous coatings, UV coatings provide an extreme-glossy finish and can deliver an abrasion and rub-resistant performance.

In order to get the best results, inks must be wax-free or UV compatible.

The results show the efficiency of the dirt removal for prints overprinted with UV cured varnish compared to non-varnished (Figs. 4a and 5b). On handsheet HP₂B_{2c} are the most dirt in classes from 0,001-0,005 mm² to 0,03-0,039 mm² (dirt count-79), and occupying a total surface

of 0,54 mm² (Fig. 4a). The dirt of the size <0,04 mm² are primarily relative to the optical properties of the handsheet and refer to the surface greyness.

These dirt are not visible to the naked eye. Only 2 dirt are larger than 0,04 mm² and its area is in spot size in 0,05-0,059 mm² and 0,08-0,089 classes.

On handsheet HP₂B₂L_{2c} are also the most dirt in classes from 0,001-0,005 mm² to 0,03-0,039 mm² (dirt count-468), and occupying a total surface of 3,22 mm² (Fig. 5b). The count of these dirt spot sizes is 8,12% higher, and the area is by 83,23% higher than for non-varnished prints. Also, in the dirt spot size greater than 0,04 mm² was

determined larger dirt count, including the class 0,30-0,39 (dirt count, from class 0,4-0,049 to class 0,30-0,39 – 32 dirt).

To complete understanding the process, the results of the first and second phases are considered. Handsheet HP₂B₂L_{2a} contains 7 dirt in a class ≥5,000 mm² with a total area of 72,079 mm² (Fig. 4b). Total dirt count in dirt spot size from 0,04-0,049 mm² to ≥5,000 mm² is 26 dirt with total area by 91,886 mm². The characteristic in this part of the process is the appearance of larger dirt, as well as a smaller number of invisible dirt with the naked eye, i.e. those in classes up to 0,04 mm².

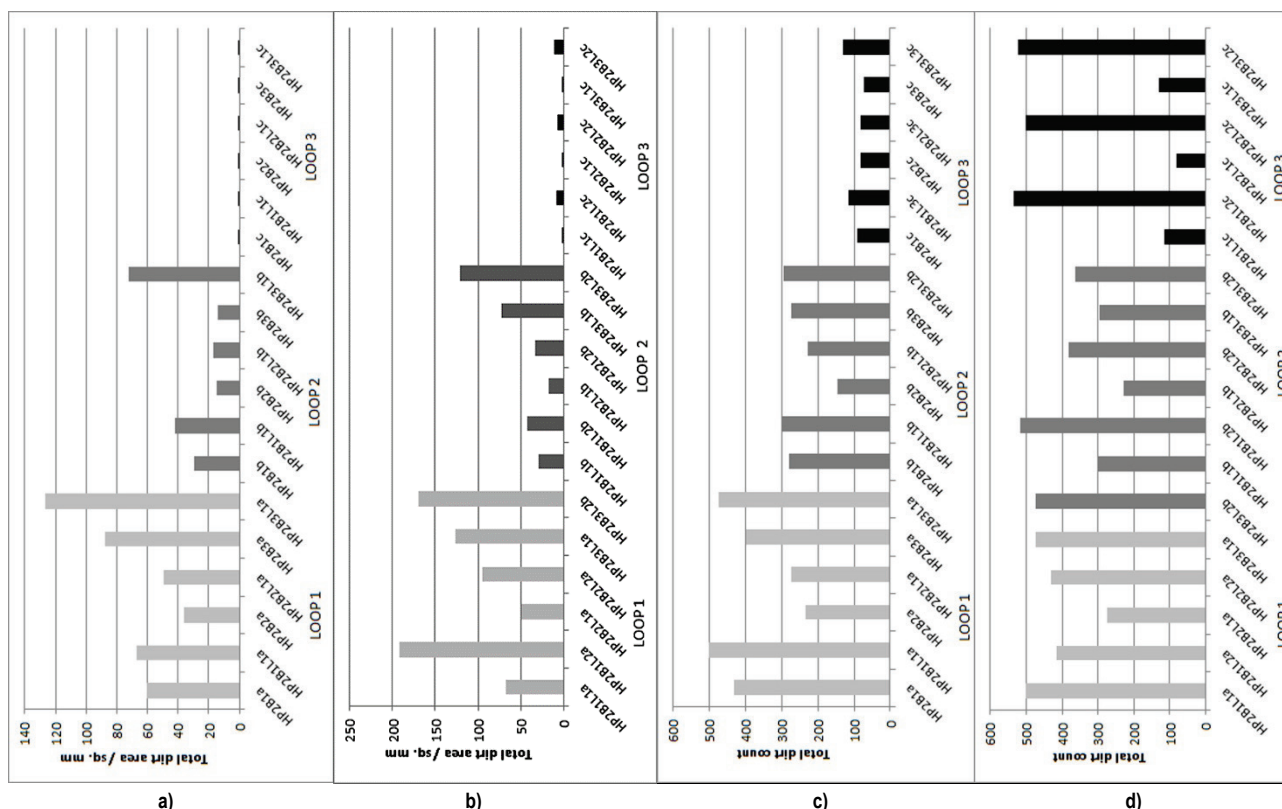


Figure 3 a) Total dirt area on handsheet versus kind of inks, prints overprinted with water-based dispersion varnish and process stages; b) Total dirt area on handsheet versus kind of inks, prints overprinted with different varnishes and process stages; c) Total dirt count on handheet versus kind of inks, prints overprinted with water-based dispersion varnish and process stages; d) Total dirt count on handsheet versus kind of inks, prints overprinted with different varnishes and process stages

Dirt Spot Size	Count	Area (sq.mm)	Dirt Spot Size	Count	Area (sq.mm)
>= 5.000	0		>= 5.000	7	72.079
4.00-5.00	0		4.00-5.00	1	4.224
3.00-3.99	0		3.00-3.99	0	
2.50-2.99	0		2.50-2.99	2	5.057
2.00-2.49	0		2.00-2.49	1	2.226
1.50-1.99	0		1.50-1.99	2	3.326
1.00-1.49	0		1.00-1.49	2	2.480
0.80-0.99	0		0.80-0.99	0	
0.60-0.79	0		0.60-0.79	1	0.694
0.40-0.59	0		0.40-0.59	1	0.502
0.30-0.39	0		0.30-0.39	2	0.686
0.25-0.29	0		0.25-0.29	0	
0.20-0.24	0		0.20-0.24	1	0.224
0.15-0.19	0		0.15-0.19	0	
0.10-0.14	0		0.10-0.14	0	
0.09-0.099	0		0.09-0.099	1	0.093
0.08-0.089	1	0.086	0.08-0.089	0	
0.07-0.079	0		0.07-0.079	0	
0.06-0.069	0		0.06-0.069	2	0.131
0.05-0.059	1	0.056	0.05-0.059	1	0.052
0.04-0.049	0		0.04-0.049	2	0.091
0.03-0.039	1	0.036	0.03-0.039	7	0.238
0.021-0.029	4	0.099	0.021-0.029	14	0.353
0.013-0.020	7	0.108	0.013-0.020	27	0.444
0.006-0.012	15	0.138	0.006-0.012	62	0.561
0.001-0.005	52	0.152	0.001-0.005	294	0.783
Totals ->	81	0.674	Totals ->	430	94.246

Figure 4 a) Distribution of dirt in 26 classes on: HP₂B_{2c} handsheet, b) Distribution of dirt in 26 classes on: HP₂B_{2a} handsheet

Dirt Spot Size	Count	Area (sq.mm)	Dirt Spot Size	Count	Area (sq.mm)
>= 5.000	2	19.586	>= 5.000	0	
4.00-5.00	1	4.093	4.00-5.00	0	
3.00-3.99	1	3.167	3.00-3.99	0	
2.50-2.99	1	2.651	2.50-2.99	0	
2.00-2.49	1	2.093	2.00-2.49	0	
1.50-1.99	0		1.50-1.99	0	
1.00-1.49	2	2.514	1.00-1.49	0	
0.80-0.99	0		0.80-0.99	0	
0.60-0.79	0		0.60-0.79	0	
0.40-0.59	1	0.489	0.40-0.59	0	
0.30-0.39	0		0.30-0.39	1	0.328
0.25-0.29	1	0.269	0.25-0.29	2	0.527
0.20-0.24	1	0.226	0.20-0.24	1	0.206
0.15-0.19	1	0.199	0.15-0.19	5	0.900
0.10-0.14	6	0.762	0.10-0.14	5	0.631
0.09-0.099	3	0.283	0.09-0.099	1	0.091
0.08-0.089	0		0.08-0.089	4	0.330
0.07-0.079	1	0.079	0.07-0.079	2	0.151
0.06-0.069	4	0.263	0.06-0.069	0	
0.05-0.059	0		0.05-0.059	7	0.367
0.04-0.049	4	0.177	0.04-0.049	4	0.176
0.03-0.039	3	0.108	0.03-0.039	10	0.323
0.021-0.029	11	0.278	0.021-0.029	14	0.346
0.013-0.020	21	0.346	0.013-0.020	35	0.579
0.006-0.012	45	0.421	0.006-0.012	74	0.672
0.001-0.005	273	0.738	0.001-0.005	335	0.903
Totals ->	383	38.742	Totals ->	500	6.529

Figure 5 a) Distribution of dirt in 26 classes on HP₂B_{2L2b} handsheet, b) Distribution of dirt in 26 classes on HP₂B_{2L2c} handsheet

On the handsheet HP₂B₂L_{2b}, is determined a smaller dirt count and area in the largest dirt spot size class $\geq 5,000$ mm² (dirt count 2, HP₂B₂L_{2b}/HP₂B₂L_{2a} = 71,43%, dirt area 19,586 mm², HP₂B₂L_{2b}/HP₂B₂L₂ = 72,63%) in comparison to loop I (Fig. 5a). All dirt is removed for dirt spot size from $\geq 5,000$ mm² to 0,40-0,59 mm² in loop III.

For the producer of recovered pulp, the quality criteria are the cleanliness and brightness of the sheet. The criteria for cleanliness are a minimum dirt count. A lot of large dirt on paper affects its mechanical properties, which can cause shooting paper roll in printing.

Significant secondary fiber quality specification of recovered pulp is also brightness. The introduction of the effective residual ink concentration measurement has given a means to relate ink content with brightness. An ink removal efficiency based on effective residual ink concentration and dirt count across the different phase of process gives the information what has happen to the ink.

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.

Brightness and the effective residual ink concentration are shown for handsheet made from process fibers (Fig. 6).

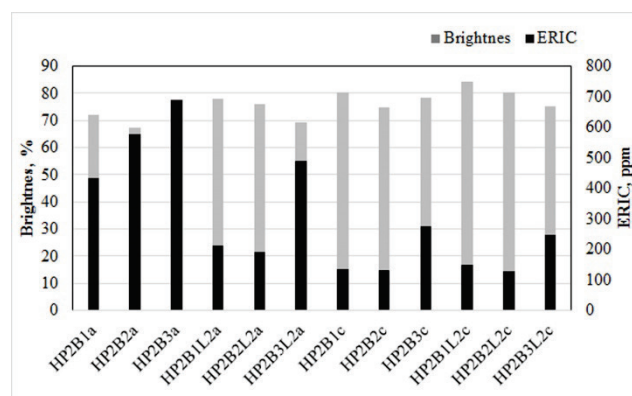


Figure 6 Brightness and the effective residual ink concentration is shown for handsheet made from process fibers

The results show that handsheets HP₂B₁L_{2c}, HP₂B₂L_{2c} and HP₂B₃L (the prints were overprinted with UV cured varnish) mainly have something higher brightness in regards to HP₂B₁, HP₂B₂ and HP₂B₃ (the prints were not overprinted with varnish). Such results are clarified by the fact, that the handsheets HP₂B₁L_{2c}, HP₂B₂L_{2c} and HP₂B₃L have a greater number of larger dirt, which cause the handsheet's optical inhomogeneity. The greater fragmentation is characteristic of the prints HP₂B₁, HP₂B₂ and HP₂B₃, which causing greyness and decreasing the handsheets brightness.

The results shown that the least amount of residual ink particles appears on a handsheet, made from fibers when in the process is used prints with ink B₂. This pattern has brightness gain HP₂B_{2c} - HP₂B_{2a} = 7,37 and Δ ERIC = HP₂B_{2c} - HP₂B_{2a} = 444,7, unlike handsheet HP₂B₂L₂ which has a somewhat worse result (brightness gain HP₂B₂L_{2c} - HP₂B₂L_{2a} = 4,18 and Δ ERIC = HP₂B₂L_{2c} - HP₂B₂L_{2a} = 61.8.

Printing technique, kind of ink, paper surface, pulping condition, mechanical action can modify the different sizes of ink particles.

Ink properties are important, since they influence ink detachment from the substrate. Removal of detachment ink particle depends on ink formulation, ink particle properties and ink particle surface. 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. These components absorb oxygen that causes them to oxidize and the cross-linking structure appears. Vegetable-based inks, especially some unsaturated vegetable oils, can cause ink detachment problems. Some resins added into the ink can cause attachment of the inks onto fibers.

Paper surface properties are also important, since they affect the ease of ink detachment. Inks printed on a coated paper surface attach more easily than inks printed directly on the fibers on an uncoated substrate. Ink detachment from the substrate is a function of the sheet surface processes and drying mechanisms. Coating dirt result from binding between the pigment and the coating material in printed coated paper.

Varnish dirt are mainly from UV cured varnish and lead to dirt of large size. Varnish cured by UV radiation, generally lead to resistant dirt that have to be fragmented.

4 CONCLUSION

Based on the results of the fibers reused from the sheet fed offset prints on cardboard made from blooms of the green algae and FSC fiber and three inks type (ink B₁ - vegetable based, contain about 80% renewable raw material, B₂ - vegetable based free of mineral oil, B₃ - mineral oil-base) the following conclusions were reached:

Compared with the mineral oil-based ink, the improved vegetable ink B₂ gives better results in terms of brightness, ink content and dirt contamination (dirt count and area) along each loop. Higher brightness gain is obtained with the newly improved vegetable based ink B₁ and good results are obtained for mineral oil based ink.

Something lower brightness gain and Δ ERIC are obtained with water-based dispersion overprint samples. The lowest results are obtained for samples overprinted with UV cured varnish. All dirt of large size are removed including dirt spot size from ≥ 5.000 mm² to 0.40-0,59 mm² (HP₂B₂L_{2c}).

An additional ecological advantage is that the flotation and accompanying chemicals have not been used in the process.

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

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