Laboratory paper pulp deinking: an evaluation based on Image Analysis, ISO Brightness and ERIC

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SUMMARY

Image analysis, ERIC and ISO Brightness measurements were used to evaluate the effectiveness of laboratory deinking assays. The accurate measurement of the residual ink amount is difficult and the results depend on the methodology used. The three techniques correlate only when the same paper pulp sample is analysed and when the ink particle size distribution profile is similar. As the relative amount of each particle size depends on the deinking protocol used, the ink removal effectiveness is measured differently according to each test method. Image analysis was shown to be the most reliable method.

KEYWORDS
paper pulps, laboratory deinking, image analysis, ERIC, Brightness

INTRODUCTION

Deinking is one of the most important stages of the paper recycling process. It involves the detachment of ink from the surface of the fibres, the dispersion of the ink particles and their subsequent removal from the mixture. The degree of deinking is affected by the chemical composition of the paper and ink, and by the ink printing method, as these determine the type of fibre-ink interactions and consequently the complexity of the ink removal process. Indeed, the development of a deinking technology requires an accurate evaluation of the characteristics of both the original pulp and the final product (*1*). It is necessary to access the pulp drainage ability, the paper strength and the amount of ink in the samples. Generally, both the physical and mechanical properties of the pulp and paper are easily determined by using well known standard procedures. The ink amount, however, is much more difficult to determine. It may be evaluated directly, by using **Image Analysis** (IA), or indirectly, by measuring **ISO Brightness** or the **Effective Residual Ink Concentration** (ERIC).

IA is based on the traditional ink-particle counting methods, which were previously quite tedious. However, automatic counting systems have made the IA method a simpler and more effective tool for the evaluation of ink, and this method is currently used by several researchers (*2-11*). Nevertheless, an accurate description of both the IA system and the operating conditions used in the evaluation are required, in order to assure the validity of the obtained results. It has been recognized that errors in the IA methods are generally related to the inaccuracies in the imaging device, to systematic errors occurring during image analysis and to the sampling procedure (*8*).

ISO Brightness is the reflectance of blue light at an effective wavelength of 457 nm. This wavelength is particularly sensitive to different characteristics of paper pulps (*12*) and it is a useful parameter that is easily interpreted relative to others that use the whole range of visible spectrum. However as the measure disregards the other portions of the visible spectrum (yellow and red) it is not expected to allow the complete characterisation of the samples.

The ERIC method evaluates the residual ink by measuring the absorbed light in the infrared range, namely at 950 nm. At this wavelength, the ink absorbs significantly more radiation than paper, and the measurements are quite insensitive to the presence of lignin, dyes or other colorants. (*13-14*).

In the research presented here, an attempt is made to establish a correlation between these three methods, and to compare their accuracy and sensitivity in the measurement of the ink amount in paper sheets.

METHODS AND MATERIALS

Paper pulps

A variety of paper pulp samples was used in the present study. These samples were prepared from: (i) **ONP**, a mixture of old-newspaper and magazines; (ii) **MOW**, a mixture of office wastepaper; (ii) **DMOW**, a deinked pulp from a mixture of office wastepapers; (iii) **MIX**, a selection of laser, inkjet and photocopy printed papers; (iv) **PHOT** and **LAS**, pure photocopy and laser paper samples, respectively. ONP, MOW and DMOW were kindly supplied by the paper company *Renova, S.A.* (Torres Novas, Portugal).

Sample preparation

In order to test and compare the different ink evaluation methods, the samples were prepared according to the following procedures:

(i) ONP and MOW were obtained by disintegrating the paper supplies at the mill, and were provided by *Renova* as high consistency pulp slurries.

(ii) DMOW was obtained by further processing MOW at the mill. The pulp was treated in the presence of sodium hydroxide and surfactant, deinked by flotation and washing, before being provided by *Renova* as high consistency pulp slurry.

(iii) MOW and DMOW were mixed in different proportions at U.M.'s laboratory facilities: 100% DMOW, 10% MOW, 20% MOW, 40% MOW, 60% MOW, 80% MOW and 100% MOW.

(iv) For the samples MIX, PHOT and LAS, the selected paper-sheets were torn in small pieces (3 by 3 cm) and disintegrated in a laboratory pulper (Lam'Deinkit, Licar S.A. – Tolosa, Guipúzcoa) according to the experimental conditions in Table 1. After disintegration, the samples were recovered by dewatering through a 200-mesh wire.

(v) The MOW pulp was treated in the presence and absence (Control assays) of chemical products and subsequently washed and/or floated in order to separate the fibres from the released ink particles. (As described in Pala et al. (*1*))

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Table 1
Operating conditions during sample preparation

PULP SAMPLES	t (min)	Consistency of the pulp in tap water (%)	T (°C)	rpm
MIX4%	15	4	25	1 500
MIX10%	15	10	58	750
PHOT4%	20	4	58	1 500
PHOT10%	20	10	58	750
LAS4%	20	4	58	1 500
LAS10%	20	10	58	750

Paper characterisation

The optical properties of paper and the amount of ink present in paper sheets were characterised as follows:

Specimen preparation

After considering several specimen preparation methods (*2*,*11*,*15*), handsheet preparation ISO 5269/1: 1979 was selected for this study as it guarantees the uniformity of the ink distribution, the sheet formation and grammage, thus improving the final results. This procedure was used in all assays, in order to allow a comparison between measurements (*11*,*15*). In the present work, handsheets of 60 g/m^2 were used (1.2 g of oven-dry pulp). This grammage was selected after testing handsheets of different weight (0.4, 0.6, 0.8, 1.2 and 2.4 g) in the IA system as it allowed the capturing of the best quality images in the IA system, the necessary opacity to accurately determine ERIC and ISO brightness (< 97%) and the benefit of using the same test piece in all evaluations. The disadvantage of using handsheets is the double-sidedness, with differences top to bottom side in smoothness, brightness and ink particle retention. (*6-8*,*15*,*16*). Thus, all the measurements (ISO Brightness, ERIC and IA) were made using the top side of the handsheets (opposite to the wire mesh).

ISO Brightness and ERIC

The optical properties of paper sheets were measured using the COLOR TOUCH™ MODEL ISO (*Technidyne*). The measurement procedures were based on standard recommendations for this instrument for paper testing. Before being analysed, the handsheets were conditioned according to ISO 187: 1990, for 24 hours.

In order to avoid the influence of optical brightening agents (OBA) on the results, the UV-portion of the radiation was excluded during the analyses by a cutoff-filter.

The ERIC value is computed via the Kubelka-Munk analysis, thus requiring the accurate knowledge of both the absorption and scattering coefficients of the mixture components (the pulp and the ink) (*13*,*17*). By default, the equipment adopts the typical coefficients for recycled newsprint. Since the absorption coefficients of both carbon black (office-paper ink formulations) and flexographic inks (newsprint) exhibit similar spectra in the range 300 to 1200 nm (*17*), the values obtained in this work are considered to be a good estimation of the true results.

With the purpose of providing an adequately opaque path, the handsheets were folded in four for opacity testing. Each quarter of the handsheet was measured for Brightness and ERIC and the final ISO Brightness or ERIC value was given as the average of the four measures. The coefficients of variation were less than 0.1% (ISO Brightness) and 1% (ERIC).

According to the standard procedures, if appropriate sampling is performed, only 5 to 10 measures in different handsheets are needed to adequately characterise a pulp batch. Considering the amount of pulp used per assay in these experiments (25g oven dry pulp), the analysis of a single handsheet was considered sufficient to provide significant results.

Each experimental condition was tested 2 to 4 times and good reproducibility was found between independent assay results. The coefficients of variation never exceeded 1% (ISO Brightness) and 6% (ERIC).

Image analysis

The IA system essentially consists of four stages: (i) observation of the sample using a magnification system; (ii) transmission of the image by a high-resolution camera to an image processor; (iii) modification of the image by the processor in order to obtain a high-contrast black and white image; (iv) identification and characterisation of the ink particles present in the image.

In the present case, the image analysis system is composed of a magnification lens (*Olympus*, model SZ-ST), an illumination device (*Olympus*, model TL2), a monochromatic CCD-camera (*Sony*, model AVC-DSCE), a CMA-D5CE adapter (*Sony*, Tokyo) and an image analysis interface DT-3152 (Marlboro, MA) (Fig. 1).

The images (40 per 60 g/m^2 handsheet) were randomly acquired using the commercial software *Image Pro Plus* 3.0 (Media Cybermetrics, Silverspring). The same magnification and lightning were used throughout the work in order to obtain comparable results (*8*,*9*,*18*).

After testing several magnifications (x10 and x20, *Diaphot* microscope, *Nikon*; x1, x2.5 and x4, *Olympus* magnification lens, model SZ-ST), a 4x objective was chosen, as a reasonable compromise between image enlargement and analysed area. It was verified that higher magnifications show a more heterogeneous fibre mat and made focusing and image acquiring more difficult, as several focal planes are obtained due to the fibre deposition in layers. Consequently, the image quality is affected and the subsequent analysis is less reliable.

The quality of the IA results is also determined by lighting. The type of light and location of the illumination relative to the analysed sheet affect the visual inter-

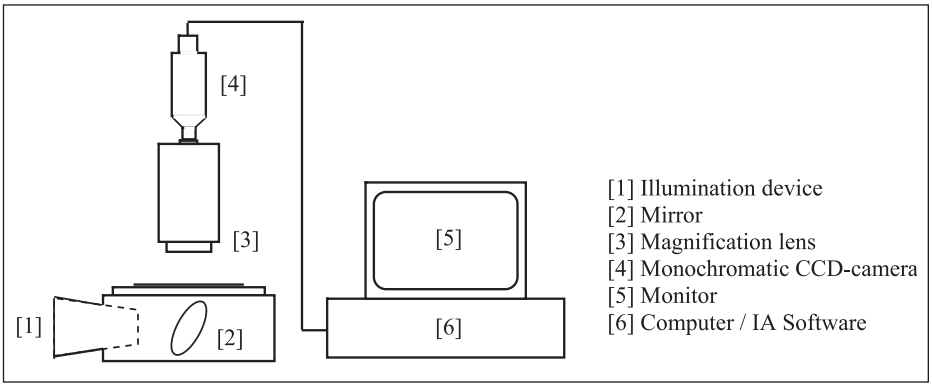

Fig. 1 Schematic representation of image analysis system

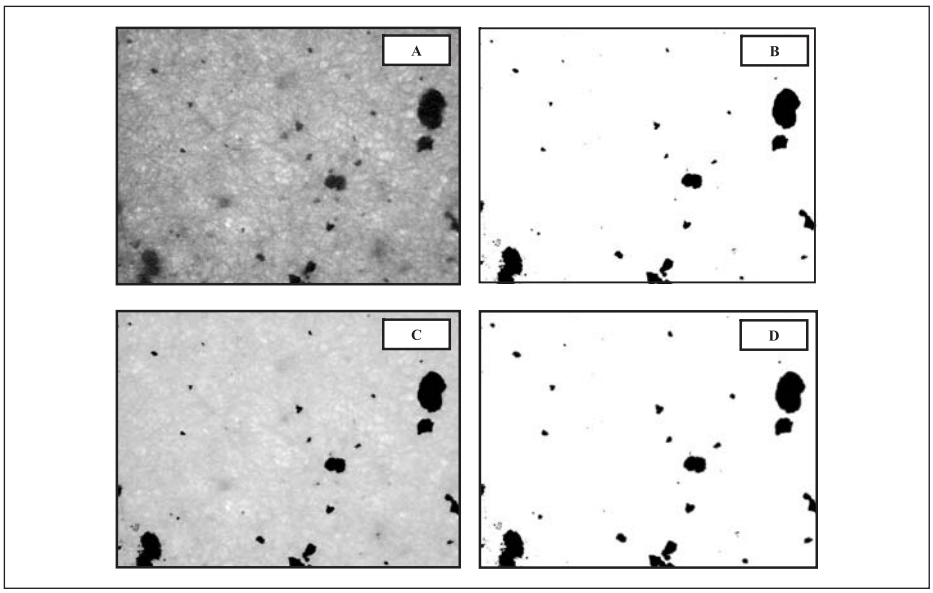

Fig. 2 Image analyse of a PHOT4% paper sample handsheet
(A) Captured image
(B) Filter tool manipulation: the effectiveness of this tool depends on the uniformity of the handsheet and the selected threshold value; non-uniform handsheets may induce background irregularities to be considered as ink particles; a low value may reduce the ink particle size thus underestimating the area covered by ink (the opposite is also valid)
(C) Captured and treated images overlapping: although some irregularities may be detected in the edge of the image, the ink particles are quite similar in both images
(D) Ink particle identification

pretation of fibres during the analysis and subsequently the fibre mat visual uniformity and the ink particle detection (*6*). In the present work, the illumination device was placed under the area of analysis and a dim mirror spread the light through the handsheets (Fig. 1). The light intensity was chosen in order to help the recognition of the ink particles: an adequate contrast between the background and the ink particles improves the image quality by highlighting the ink particle contours and reducing the shadows in the background that could cause fibre agglomerates to be identified as ink particles.

Particle counts, shapes and sizes were examined using commercially available software (*Globalab Image 3.2.*, Data Translation, Marlboro). First, the captured image was divided into a matrix of picture elements (pixels) and the light intensity of each one was evaluated. The images were then manipulated by using a filter tool which eliminates the background (converts it to white) and renders the ink particles totally black (Fig. 2). The selection of a suitable threshold value allowed the identification of the contaminants and to ensure the reproducibility of the image analysis this value was conserved throughout the work. Whenever a significant ink particle modification was detected after image manipulation, either as a clear reduction or increase in the ink particle size, the area where the problem occurred was discarded. Particles next to the image border were ignored during counting.

The IA system required calibration. The analysed area, corresponding in each image to 438 528 pixels, had an area of about 13 mm^2. The total area analysed in each handsheet was about 5.2 cm^2. The dimension of the smallest detectable particle was 297 μm^2 (10 pixel), equivalent to a diameter of 19 μm, assuming a spherical geometry for the particle. The IA system resolution is 29.86 μm^2/pixel.

Microsoft® Excel 2000 was used to perform the statistical analysis of each 40-image-set.

In order to validate the IA results, the effect of random sampling errors was evaluated by using the correlations suggested by Zeyer *et al.* (*8-9*). These equations associate the relative error of measured impurities with the analysed area, the average particle size and the level of impurities in the range 10 to 1000 ppm, thus requiring previous testing for application within higher impurity levels (as in the present work). The comparison between both the standard deviation (SDEV) and the confidence interval for a confidence level of 95% (CI), obtained after indirect determination, by using the ink particle characteristics, and after direct determination, by using the experimentally detected total area covered by ink, demonstrated a good correlation between these statistical parameters, thus indicating that the Zeyer approach could be applied to higher levels of dirtiness in the samples (Table 2). In the present work, the greatest SDEV and CI (8-9) obtained, were 14% and 8%, respectively. Moreover, the coefficient of variation between equivalent assays (same experimental conditions) never exceeded 10%.

RESULTS AND DISCUSSION

ISO Brightness, ERIC and IA of MOW and DMOW mixtures

Mixed-office wastepaper handsheets containing different amounts of ink were analysed. These handsheets were obtained by mixing MOW and DMOW pulps in different proportions. The ink

Table 2
SDEV and CI for the total area covered by ink in MOW (experimental data statistical analyse versus Zeyer et al.) *

Sheet	A_M	SDEV (A_i)	Ink area (ppm)
1	2 364	3 284	10 714
2	2 453	3 506	10 080
3	2 332	3 672	10 031
4	2 492	3 542	10 325
	2 408	3 496	10 291
% SDEV (experimental) **			3.0
% CI (experimental) **			3.0
% SDEV (predicted) ***			4.3
% CI (predicted) ***			2.7

* The evaluation considered the analysis of 3.9 cm^2 per sheet (equivalent to the acquisition of 30 images); A_M, ink particle average size; A_i, individual ink particle size.
** SDEV and CI for the total area covered by ink (Excel 2000 statistical analyses of the experimental ink area values)
*** SDEV and CI for the total area covered by ink (Zeyer *et al.*, 1995a, 1995b evaluation (*8,9*))

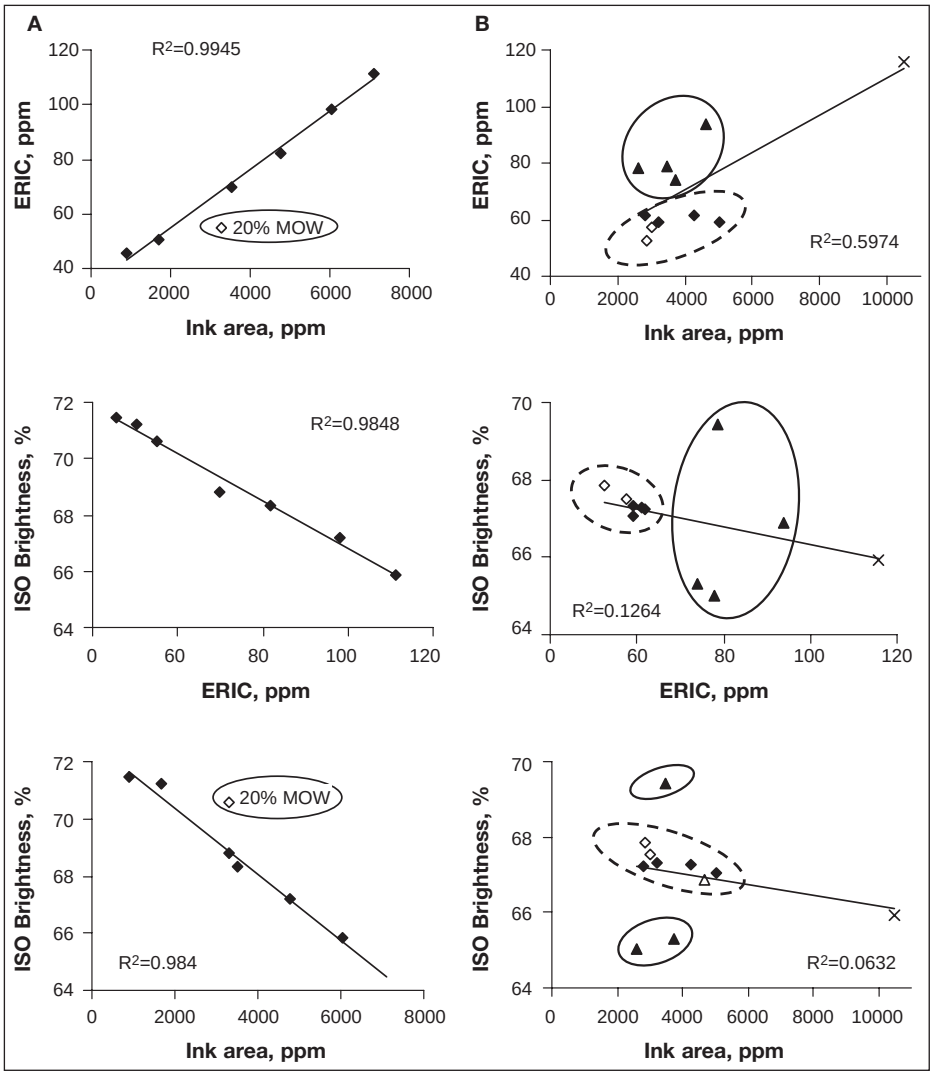

Fig. 3 Correlation between residual ink evaluation methods
(A) MOW/DMOW mixtures
(B) Chemically deinked MOW samples: X, Non-treated ♦, Washed samples ◊, Floated+Washed samples ▲, Floated samples Δ, Floated samples present in the washed samples tendency area ⬭, Floated samples trend ⬭, Washed samples trend

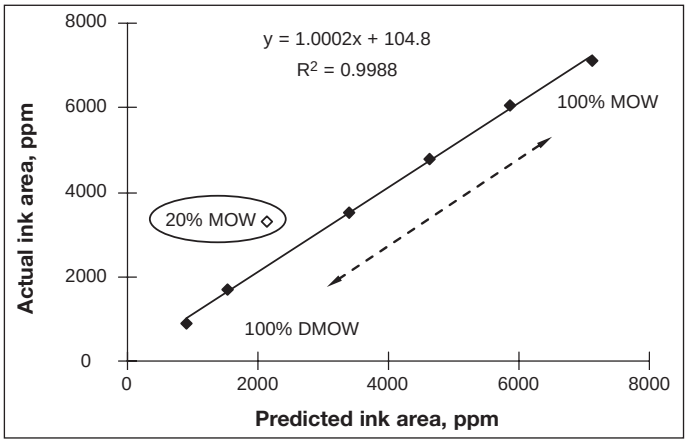

Fig. 4 Actual ink concentration versus predicted ink concentration by IA (MOW and DMOW mixtures) The "predicted ink area values" were calculated using the ink area values obtained in pure MOW and DMOW and the amount of each of these pulps in the intermediate samples.

concentration in each MOW/DMOW mixture was evaluated by using IA, ISO brightness and ERIC (Fig. 3). Moreover, considering the IA results of the 100% MOW and 100% DMOW samples, the "predicted" ink concentration for the 10 to 80% MOW handsheets was estimated in order to evaluate the IA results (Fig. 4).

According to Zeyer et al. (*8*), whenever the same equipment and the same settings are used to perform all the IA measures (as in this work), the sampling procedure is the major factor in obtaining valid IA results. As the contaminant distribution on the pulps occurs randomly, it is necessary to assure that an adequate amount of sample is analysed. In the present work, the IA of the MOW/DMOW mixtures showed that the cleaner the pulp, the higher the SDEV and the CI related to the total area covered by ink (MOW: 4.2% SDEV and 2.5% CI versus DMOW: 13.2% SDEV and 7.2% CI). Indeed, if the same area of analysis is considered and the average ink particle size in the samples is similar (MOW ≈ 1977 μm² *versus* DMOW ≈ 2043 μm²), a lower particle count (MOW ≈ 1278 particles ≈ 7113 ppm *versus* DMOW ≈ 157 particles ≈ 903 ppm) explains the higher statistical error. A similar effect was detected when samples containing larger ink particles and a very dissimilar ink particle size population were analysed under the same IA operating conditions (e.g. MIX4% and MIX10% samples, data not shown). Apparently, in these cases, a higher paper area should have been analysed in order to reduce the error on the IA measure. Nevertheless, the correlation shown on Figure 4 (MOW/DMOW mixtures) seems to indicate that the settings that were established to perform the IA in the present work permit an accurate ink area measure. According to the figures, the "predicted" ink concentration correlates well with the "actual" ink concentration. In fact, only the 20% MOW value deviates from the linear correlation, probably because of a non-identified experimental error. Moreover, regarding the low amount of pulp (≈ 25g oven dry) used in the laboratory deinking assay evaluations (*1*), it is believed that the selected sampling area (5.2 cm²) provides valid results. Other authors describe IA areas lower than 5.2 cm² in their studies (*11*,*16*). Indeed, reducing the area of analysis is interesting, as it saves time.

Considering the ISO brightness and ERIC measurements, it would be expect-

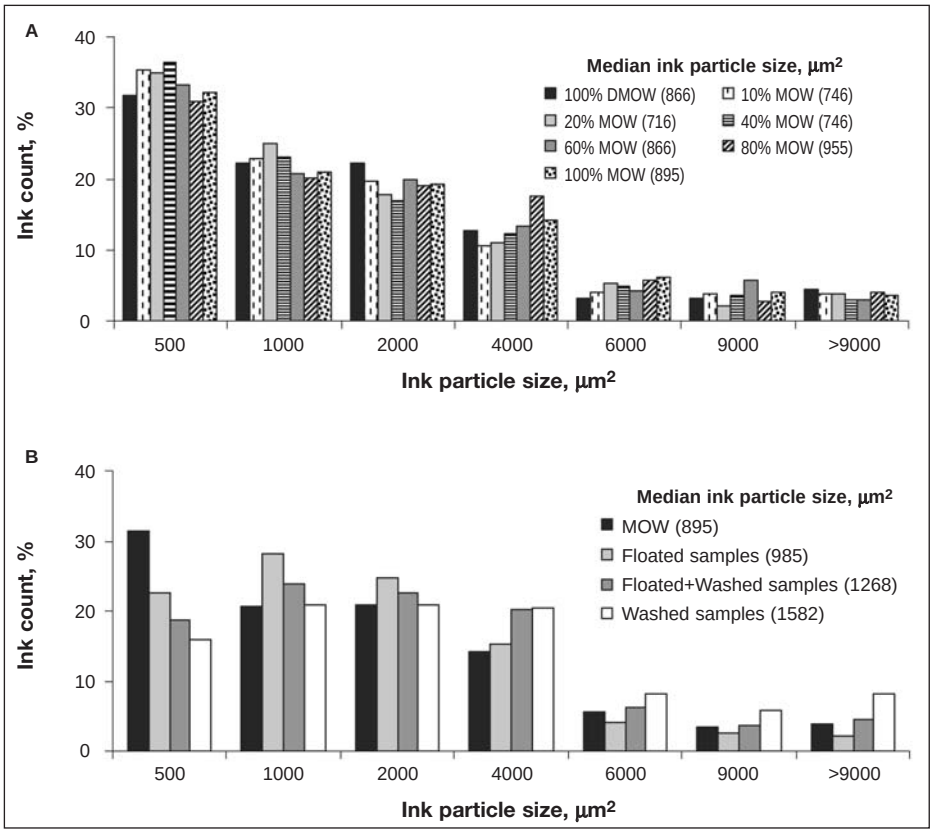

Fig. 5 Ink particle size distribution profiles
(A) MOW/DMOW mixtures
(B) Chemically deinked MOW samples

ed that an increase in the amount of ink would decrease the ISO brightness and increase the ERIC value of the paper. This effect was detected in the present work in the MOW/DMOW mixtures and the measurements correlate well with the IA results (Fig. 3). However, it is known that there is no direct relationship between the ink particle concentration and the optical properties of paper (*10,19*). In fact, the type of ink, the ink particle size and the ink particle distribution in the paper all make a major contribution to the result. Furthermore, ISO brightness is a macroscopic measure of reflectance and in consequence it measures the contribution of all the other pulp constituents in addition to the ink: fibres, additives and chemicals (*2,20*). In this work, the good correlation between ISO brightness, ERIC and ink particle concentration (IA) found in the MOW/DMOW mixtures is most likely attributed to the similarity of the ink particle size distribution profile in all the analysed samples (Fig. 5) and to the similar paper pulp composition.

ISO Brightness, ERIC and IA of different paper pulp samples

The dirt content of samples obtained by disintegrating different paper furnishes was also evaluated. As shown in Table 3, it was not possible to measure the ISO brightness and the ERIC in the PHOT and LAS samples. This is probably due to the presence of optical brightness agents (OBA) in the pulp, which are often added to printing/writing paper in order to improve their visual appearance and quality. Although the optical analysis of paper was carried out in the absence of UV radiation, it is known that the OBA may also be excited by the short-wavelength visible radiation (400 to 440nm) (*14*), and this radiation was not excluded during these measurements. When this light strikes the paper it is absorbed by the fluorescent material and re-emitted at a longer wavelength in the blue portion of the spectrum, thus increasing the reflectance value at 457nm. Whenever this value is higher than the total reflectance of the sample, the ISO brightness and ERIC cannot be determined (*21*).

Even when the optical methods provide measurable values, as in the MOW, ONP and MIX samples, a careful analysis is necessary in order to evaluate their use as a dirtiness measure. The pulp manipulation during the recycling process does not remove the majority of the OBA added to the pulps during the previous production cycle (*22*). In addition to the effect of the OBA, some of the chemical products that are used in the deinking process (such as hydrogen peroxide and sodium hydrosulfite) may contribute to the increase of the residual fluorescence of the pulp (*22-23*). The subsequent increase of reflectance in the blue portion of the spectrum causes the paper to appear whiter to the observer, but this is not necessarily related to a cleaner pulp.

MOW, ONP and MIX handsheets analysis reveals no correlation between the ISO brightness, the ERIC and the IA

Table 3
Dirtiness evaluation by IA, ISO brightness and ERIC on different pulp samples

	MOW	DMOW	ONP	LAS		PHOT		MIX	
				4%	10%	4%	10%	4%	10%
Total ink area (ppm)	7 113	903	4 073	18 120	13 595	17 330	14 976	16 941	8 615
Particle count (IA)	1 278	157	1 419	366	335	981	861	460	303
Minimum ink particle size (µm²)	249	239	149	298	298	298	298	298	298
Maximum ink particle size (µm²)	37 726	27 041	29 250	340 910	203 734	252 444	194 870	426 898	214 270
Ink particle size (µm²)	1 977	2 043	1 020	20 692	15 202	6 466	6 356	19 265	14 870
Median (µm²)	895	866	478	4 880	6 029	2 089	2 119	2 731	4 477
ISO brightness (UVEX) *, %	65.84	71.45	58.41	**	**	**	**	73.5	68.3
ERIC (UVEX) *, ppm	111.3	45.7	190.2	**	**	**	**	77.9	78.2

* UVEX, UV-portion of the radiation excluded
** ISO brightness and ERIC were not determined probably due to the presence of optical brightening agents in the pulps

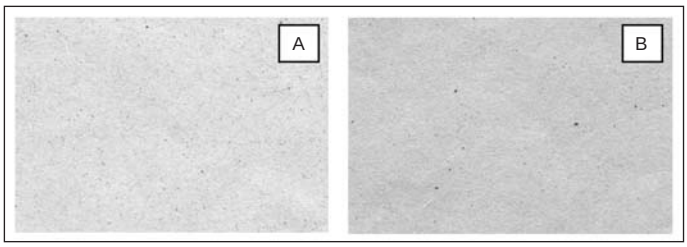

Fig. 6 Visual assessment of MOW (A) and ONP (B) paper sheets

values (Table 3). However, another effect is exposed by these measurements: whenever the ink is finely divided in paper, a loss of ISO brightness and a gain in ERIC occurs. This can be seen by comparing ONP with the other pulps. ONP contains the smallest ink particles (1020 μm^2) and presents the lowest area covered by ink (4073 ppm); it has the highest ERIC and the lowest ISO brightness values (58.4% ISO and 190.2 ppm ERIC) of the three pulps. In turn, MOW contains 1977 mm2 ink particles, an area covered by ink of 7113 ppm, 65.8% ISO and 111.3 ppm ERIC. Similar results were obtained by other authors (*13*). This effect is probably due to the fact that whenever the ink is broken down into smaller particles, and is distributed evenly throughout the paper sheet, each individual ink particle has its light absorbing characteristics nearly maximized. On the contrary, when the ink is agglomerated in large blobs, most of the ink particles do not have the capacity for absorbing light because they are surrounded by other ink particles, thus justifying the ERIC decrease and the ISO brightness increase (*13*,*21*).

Figure 6 highlights the effect of the ink particles distribution in the visual assessment of the paper. Although it contains less ink, the ONP sample (ink particle median size ≈ 478 μm^2) seems darker to the observer than the MOW sample (ink particle median size ≈ 895 μm^2). According to some authors the ERIC analysis does not represent a measure of the ink amount present in the paper sheets, but instead measures the visual effect that the residual ink produces in the paper (*13*). The ERIC values obtained in this work appear to corroborate this view (ONP ≈ 190.2 ppm versus MOW ≈ 111.3 ppm).

The usefulness of the ISO brightness measurements may also be affected by the fact that the ink absorbs mainly in the infrared portion of the spectrum (800 to 1200 nm). In consequence, it might happen that no significant differences are detected during the brightness analysis (in the 400 to 500 nm range) no longer reflecting the amount of ink in the paper (*13-14*). In fact, it is possible that two sheets that are visually different to the human eye, can present the same ISO Brightness value. (This situation, however, was not detected in the present work.)

In comparison to the other techniques, image analysis has the great advantage of giving a direct measure of the ink amount, allowing a detailed characterisation of the ink particles. The values in Table 3 reveal the modification of three types of paper (LAS, PHOT and MIX) during disintegration at different consistency and/or temperature. This type of characterisation, can be important in practice, allowing the optimisation of the pulping stage and the selection of appropriate subsequent deinking stages, as the ink particle characteristics affect the effectiveness of the cleaning processes (*16*,*19*,*24-26*). Regarding printed ink, a higher consistency promotes ink film fragmentation, specially in the LAS and MIX samples, where the average ink particle size is reduced by 26% and 22%, respectively (Table 3). The lower number of particles and ink area values at the 10%-consistency also indicates a more extensive detachment of the ink, and subsequent ink loss during the pulp dewatering and fibre recovering step.

According to the statistical parameters (Table 3), the extent of the fragmentation/detachment of the ink depends on the type of printing (e.g. LAS versus PHOT). Considering that the inks used in laser and photocopy prints are usually made of 55 to 90% thermoplastic polymers and of 40 to 50% pigments and additives, water does not interfere in the structure of the printed film (*27-28*). The ink film modification is mainly caused by the mechanical action on the fibres, the swelling of the fibres and the temperature of the operation (*26*,*28-29*). Consistent with these observations, it appears that PHOT might contain polymers that are less elastic at the disintegration temperature than the ones in LAS or might include a thinner and/or less dense printed xerographic film, thus explaining the more brittle film (*30*). In MIX however, the dispersion on aqueous medium may also be occurring to a limited extent because it contains some ink jet prints, which are soluble in aqueous solvents.

At high temperatures the thermoplastic polymers are softened and the ink films adhere less strongly to the fibres, allowing an easier detachment of the ink as the result of a more effective mechanical action. If the 55°C to 58°C temperature range during the MIX disintegration at 10% includes the glass-transition temperature of the inks in MIX (usually, 50 to 70°C (*31*)) this may be another explanation for the improved detachment of the ink in MIX10%. The ink softening at high temperatures also contributes to the presence of a higher amount of totally "clean" ink particles in the suspension (*26*).

In all assays, a sphericity coefficient was measured by IA in order to analyse the influence of the disintegration conditions on the ink particles shape. Neither the consistency nor the temperature produced a significant change in this coefficient: LAS (0.8); PHOT (0.9); MIX (0.8).

ISO Brightness, ERIC and IA of chemically deinked samples

Deinking was evaluated by comparing the ink amount on both chemically-treated and non-treated pulp samples. In contrast to the analyses of blank MOW and DMOW samples (described in the first section of Results and Discussion), this study shows that the correlation between the optical parameters and the IA does not exist for pulps subjected to different deinking sequences (Fig. 3). This is probably due to the modification of the ink particle size distribution profiles of the pulps during the deinking procedure (Fig. 5). As is well known, the effect of retention of the ink particles in the fibre mat makes the washing step adequate for the removal of small particles (1 to 10 μm), whereas the probability of collision between the ink particles and the air bubbles (and consequent attachment and formation of the ink particle – air bubble complex) makes the flotation step more efficient for the removal of the larger ones (10 to 150 μm) (*32-35*). Indeed, the ink particle median size of the samples is higher after washing than after flotation thus indicating a higher removal of the

Table 4
Dirtiness evaluation by IA, ISO brightness and ERIC on deinked and non-deinked MOW samples

	MOW	FLOATED+WASHED SAMPLES		FLOATED SAMPLES		WASHED SAMPLES	
		Control	Assay	Control	Assay	Control	Assay
Total ink area (ppm)	7 113	3 018	2 864	6 589	3 728	5 030	3 220
ISO brightness (UVEX), %	65.84	67.5	67.9	65.0	65.3	67.2	67.3
ERIC (UVEX), ppm	111.3	57.5	52.5	78	74	61.7	59.1

smaller ink particles during the first cleaning step (Fig. 5). As stated before, the ISO brightness and the ERIC values are affected by the ink particle size, particularly by the smaller sizes. This effect may make these measures quite insensitive to the ink removal, thus hindering the ink removal effectiveness. This situation is evidenced by the results obtained after using the three different ink removal protocols on the control MOW samples, namely the flotation and/or washing (Table 4). Indeed, the IA evaluation shows more significant changes in the pulp dirtiness than the ISO brightness or ERIC measurements do. Moreover, the ISO brightness values reveal a higher spread in the results whenever the amount of smaller particles is higher (namely, on the samples subjected to flotation) (Fig. 3). Actually, due to the effect of small particles on ISO brightness, the measured variation in the control/assay samples, already reported, was minimal although the different amounts of dirt present in the samples could be seen by the naked eye (*19*,*36*).

Considering the ERIC and IA results it appears that, although there is a lack of correlation between these two measurements, it is possible to use either method in order to evaluate the deinking process. However, the differences between the techniques (Table 4) is a major limitation when comparing different research results in this area. According to the results the ERIC/IA correlation seems to improve when similarly treated samples are considered (Fig. 3). The flotation and the washed samples tend to establish two different trend areas in the graphics presented in Figure 3.

CONCLUSIONS

An accurate evaluation of a deinking system performance requires the use of reliable measures and sensitivity to small dirt particles, as well as expeditious methods. According to the present work, IA and ERIC may be used as deinking evaluation tools, but different properties are assessed. On the contrary, ISO brightness does not always reflect the changes ocurring in the samples and shows a significant lack of specificity in the quantification and characterisation of the contaminants. In fact, the correlation between the results is possible only if the ink particle size distribution profiles are similar in all analysed samples.

ERIC evaluation is faster and simpler than the IA but is highly influenced by the presence of OBA agents and small ink particles. Moreover, as it represents a measure of the visual aspect of the paper, ERIC only allows a comparative assessment of the samples. On the other hand, as IA is related to a direct identification and characterisation of the objects (ink particles), it gives an accurate measure of the ink amount. Furthermore, it is not affected by the other paper contaminants thus allowing evaluation over all sorts of samples. However, IA measures are dependent on a wide range of experimental factors, so it is imperative to validate the results by calibrating the IA system.

When applied correctly, IA is also very useful because it provides information that can help deinking optimisation by indicating what is happening during the early stages. In the present work, it has been shown that the sample preparation stage can be controlled in order to favour the detachment and limit the fragmentation of the adhered ink film. Generally, high consistencies and mixing must be restricted in order to avoid ink fragmentation and redeposition on the fibres; high temperature softens the printed ink and favours its detachment from the fibres surface. This processing changes the appearance of the ink particles, namely the size. These modifications will have important effects in subsequent deinking stages.

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REFERENCES

(*1*) Pala, H., Mota, M. and Gama, F.M. – Enzymatic versus chemical deinking of non-impact ink printed paper, *J. Biotechnol.* **108**(1):79 (2004).

(*2*) McCool, M.A. and Taylor, C.J. – Image analysis techniques in recycled paper, *Tappi J.* **66**(8):69 (1983).

(*3*) McCool, M.A. and Silveri, L. – Removal of specks and nondispersed ink from a deinking furnish, *Tappi J.* **70**(11):75 (1987).

(*4*) Zabala, J.M. and McCool, M.A. – Deinking at Papelera Peninsular and the philosophy of deinking system design, *Tappi J.* **71**(8):62 (1988).

(*5*) Klungness, J.H., Fernandez, L.E. and Plantinga, P.L. – Image analysis for measuring adhesive contaminants in pulp, *Tappi J.* **72**(1):89 (1989).

(*6*) Jordan, B.D., Nguyen, N.G. and Trépanier, R.J.R. – Measuring the particle-size distribution of residual ink in recycled paper, *Tappi J.* **76**(10):110 (1993).

(*7*) Stuman, F.J. – Sampling statistics applied to automated TAPPI dirt counting, *Tappi J.* **77**(5):179 (1994).

(*8*) Zeyer, C. Venditti, R.A., Puangchinda, K.W. and Heitmann, J.A. – The distribution of impurities in pulp and paper: the effects of the random distribution of impurities on image analysis, *Tappi J.* **78**(2):168 (1995a).

(*9*) Zeyer, C. Venditti, R.A. and Heitmann, J.A. – The effect of sample size on the confidence interval for image analysis of impurities in pulp and paper, *Tappi J.* **78**(2):176 (1995b).

(*10*) Riviello Jr., A.E., Scamehorn, J.F., Christian, S.D. and Borchardt, J.K. – The use of image analysis to measure brightness of paper, *TAPPI Pulping Conference Proceedings*, p.165 (1995).

(*11*) Lévesque, M., Dessureault, S., Carabin, P. and Barbe, M.C. – Measurement of deinked pulp quality and deinking efficiency, Part II: Impact of specimen preparation procedures on ink content results, *Pulp Paper Can.* **99**(6):79 (1998).

(*12*) Clark, J. D.'A. – **Pulp Technology and Treatment for Paper,** 2nd ed., Miller Freeman Publications Inc., San Francisco, USA (1985).

(*13*) Jordan, B.D. and Popson, S.J. – Measuring the concentration of residual ink in recycled newsprint, *J. Pulp Paper Sci.* **20**(6):J161 (1994).

(*14*) Popson, S.J., Malthouse, D.D. and Robertson, P.C. – Applying brightness, whiteness, and color measurements to color removal, *Tappi J.* **80**(9):137 (1997).

(*15*) Dorris, G.M. – In detachment and flotation efficiency in ONP/OMG blends. Part I: Effect of specimen preparation procedure in estimates, *J. Pulp Paper Sci.* **25**(1):1 (1999).

(*16*) Prasad, D.Y., Heitmann, J.A. and Joyce, T.W. – Enzyme deinking of black and white letterpress printed newspaper waste, *Progress in Paper Recycling* **1**(3):21 (1992).

(*17*) Jordan, B. and O'Neill, M. – The Kubelka-Munk absorption coefficients of several carbon blacks and water-based printing inks, J. *Pulp Paper Sci.* **20**(12):J371 (1994).
(*18*) Moss, C.S. – Image analysis standardization, *Tappi J.* **76**(10):117 (1993).
(*19*) Prasad, D.Y., Heitmann, J.A. and Joyce, T.W. – Enzymatic deinking of colored offset newsprint, *Nordic Pulp and Paper Research J.* **2**:284 (1993).
(*20*) Gigac, J., Ku_a, V. and Schwartz, J. – Effects of fibres and fillers on the optical and mechanical characteristics of paper, *Tappi J.* **78**(2):162 (1995).
(*21*) COLOUR TOUCH™ MODEL ISO Instruction Manual, *Technidyne Corporation*, USA (1994).
(*22*) Earl, P.F. and Znajewski, T. – Removal of fluorescence from recycled fibre using chlorine dioxide, *Pulp Paper Can.* **101**(9):62 (2000).
(*23*) Shrinath, A., Szewczak, J.T. and Bowen, I.J. – A review of ink-removal techniques in current deinking technology, *Tappi J.* **74**(7):85 (1991).
(*24*) Zeyer, C., Joyce, T.W., Heitmann, J.A. and Rucker, J.W. – Factors influencing enzyme deinking of recycled fiber, *Tappi J.* **77**(10):169 (1994).
(*25*) Snyder, B.A. and Berg, J.C. – Effect of particle size and density in flotation deinking of electrostatic papers, *Tappi J.* **77**(7):157 (1994).
(*26*) Johnson, D.A. and Thompson, E.V. – Fiber and toner detachment during repulping of mixed office waste containing photocopied and laser-printed paper, *Tappi J.* **78**(2):41 (1995).
(*27*) Bennington, C.P.J., Sui, O.S. and Smith, J.D. – The effect of mechanical action on waste paper defibering and ink removal in repulping operations, *J. Pulp Paper Sci.* **24**(11):341 (1998).
(*28*) Wielen, L.C.V., Panek, J.C. and Pfromm, P.H. – Fracture of toner due to paper swelling, *Tappi J.* **82**(5):115 (1999).
(*29*) Kakogiannos, A., Johnson, D.A. and Thompson, E.V. – Laboratory high-consistency dispersion studies of laser-printed office copy paper, Part II, *Tappi J.* **81**(4):159 (1998).
(*30*) Dorris, G.M. and Sayegh, N.N. – The role of print layer thickness and cohesiveness on deinking of toner-printed papers, *Tappi J.* **80**(4):181 (1997).
(*31*) Quick, T.H. and Hodgson, K.T. – Xerography deinking - a fundamental approach, *Tappi J.* **69**(3):102 (1986).
(*32*) Harrison, A. – Flotation deinking is critical in unit process method of deinking, In Ken L. Patrick (ed) **Paper Recycling – Strategies, Economics, and Technology,** Pulp & Paper Focus Book, Miller Freeman, San Francisco, p.144 (1991).
(*33*) Horacek, R.G. and Jarrehult, B. – Chemical application expands in washing/flotation deinking systems, In Ken L. Patrick (ed) **Paper Recycling – Strategies, Economics, and Technology,** Pulp & Paper Focus Book, Miller Freeman, San Francisco, p.139 (1991).
(*34*) Ferguson, L.D. – Deinking chemistry: part 2, *Tappi J.* **75**(8):49 (1992).
(*35*) Heindel, T.J. – Fundamentals of flotation deinking, *Tappi J.* **82**(3):115 (1999).
(*36*) Prasad, D.Y. – Enzymatic deinking of laser and xerographic office wastes, *Appita* **46**(4):289 (1993).

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