VARIOUS VALORISATION ROUTES OF PAPER INTENDED FOR RECYCLING – A REVIEW

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Dedicated to Acad. Bogdan C. Simionescu
on the occasion of his 70th anniversary

This review outlines the possibility of utilizing paper intended for recycling in different processes. Paper is mainly recycled for producing recycled paper, but in the last decade it has been also used for obtaining cellulose derivatives and bioethanol. The production of recycled fibres from paper by the conventional chemical flotation deinking process is the most widely used method worldwide and therefore, the most often described. In addition to flotation, the methods involving enzymes, ultrasound and adsorbents are also promising, but their application in paper recycling facilities is still limited. The efficiency of these processes mostly depends upon the interaction between papers and printing inks. The presence of printing ink residues in recycled pulp can cause health and safety problems because of their toxic components. Moreover, the effluents from paper recycling plants are influenced as well by the raw materials used for recycling, which may lead to high organic loads. The paper collected for recycling is sometimes contaminated with food or is wet, and in such cases, it is recommended to use the paper for bioethanol production and organic recycling. Energy recovery *via* pyrolysis, gasification and combustion is also recommended, but only in the case the produced heat is used for other applications. In order to choose the most effective waste management method, life cycle assessment (LCA) can be used. However, the existing published literature generally overlooks the aspects related to the presence of printing inks in the paper intended for recycling. This literature review highlights the fact that the influence of the most common non-fibrous materials in paper, which are printing inks, on the process efficiency of the recycling methods is poorly described.

Keywords: paper for recycling, printing inks, material and organic recycling, recycled fibres, bioethanol, cellulose derivatives, energy recovery, life cycle assessment

INTRODUCTION

Municipal solid waste (MSW) is the most complex solid waste stream, consisting of food residues, paper and board, plastics and other components. Paper and board, altogether with kitchen waste, are one of the most common materials in municipal solid waste and the waste material fraction with the highest recycling rates. In 2016, 72.5% of all the paper consumed in Europe was recycled.[1] Recovered paper is today the most important raw material for the production of paper, paperboard and corrugated board. The production in the packaging sector is continually increasing, whilst graphic paper (newsprint, printing and writing paper) has maintained its recent decline. CEPI members produced 90.9 million tonnes of paper and board in 2016, while the utilisation of paper and board was of 47.8 million tonnes, with a utilisation rate of 52.6%.[1] Not all the paper is recycled in the same amounts: corrugated and kraft grades, as well as newspapers and magazines, constitute more than 70% of all the recovered paper in Europe (Table 1). Most of the paper-based products mostly have a life span of a few days (*e.g.* newspapers) or a few weeks (*e.g.* packaging). Paper waste comprises not only discarded newspapers, office papers and packaging paper waste, but also the production waste from the paper industry. Different processes in the pulp and paper industry (pulping, deinking unit operations and wastewater treatment) result in the formation of solid wastes and sludge. Their amount and composition depend mostly on the paper grades produced, the raw materials used, the process techniques applied and the paper properties that have to be achieved.[2] The sludge generated from paper production and recycling processes contains mainly short fibres, coatings and fillers, such as kaolin, talc, calcium carbonate and clays, which are added to improve the finished properties of the paper products. Additionally, the sludge contains ink particles, extractive substances and de-inking additives.[3] The paper intended for recycling is mostly used as a raw material by the paper industry for the production of recycled paper and cardboard products. Sometimes, the quality of recycled paper is far poorer than that of the paper made from virgin pulps, because of the shorter fibre length and reduced tensile strength.[4] In addition, the recycled pulp sometimes contains toxic substances originating from printing inks, which are undesirable for health and safety reasons.[5] Thus, the utilization of paper intended for recycling for producing value-added materials, such as bioethanol, different cellulose derivatives, biogas and eco-composites, has been increasing all over the world in recent years. It can provide an alternative to paper recycling and serve as a way to possibly address the issues arising from paper recycling.[6,7]

Ervasti *et al*.[8] have concluded in their literature review that there is no uniform system of terms and definitions related to paper recycling. Terms such as "discarded paper", "paper for recycling", "paper stock", "refuse paper", "recovered paper", "scrap paper", "secondary paper", "used paper" and "waste paper" have been used to define the same product. In addition, the term paper often refers to both paper and paperboard. Thus, they suggest that all authors should always define these terms when they use them in a study. In addition, there are some differences between the uses of the term "recovery", which is defined differently in the United States (to include only collection) and in Europe (to include reuse, material recycling, composting and energy recovery).[8] Thus, in this paper, the term "paper for recycling" will be used.

Paper collected for recycling to be valorized in some of the proposed ways is usually not clean and very often it contains different impurities, such as printing inks, coatings, or even residues of food, when it comes to food packaging. Therefore, the aim of this study is to provide an overview of the influence of the most significant components of paper for recycling, such as printing inks, on the efficiency of the waste management methods.

COMPOSITION AND QUALITY OF PAPER FOR RECYCLING

Paper and paper products are made primarily of mechanical and/or chemical wood pulp, recycled fibres, non-fibrous components (minerals and additives) and water.[9] Fibres are made of cellulose, hemicellulose and lignin. Additives and fillers guarantee particular properties of the papers. Many papers are coated or partly varnished to improve the surface properties. As presented in Table 2, different paper products will have different composition. Office paper contains lower amounts of lignin, compared to other types. Higher concentrations of metals (Al, As, Ca, Cd, Cr, Cu, Hg, Pb, Sb, Sn, Nd) in paper and cardboard waste may originate from printing inks and coatings,[10–12] while C, H and Al originate from materials included in cardboard and paper composites (plastic and aluminium layers). The highest ash contents can be found in magazines, printed advertisement and books that can be associated with higher Ca contents due to fillers (calcium carbonate and kaolin) in these fractions.[10]

Paper for recycling can also contain different materials, such as staples, laminated covers, plastic wrapping, inks, thick adhesive layers *etc*.[13] In general, printing inks are the most important non-fibre components that can be found in or be part of the paper for recycling. In addition to printing inks, lacquers or overprint varnishes, which are uncoloured forms of printing inks used to give added gloss and protective properties to the print and substrate, can be also found as non-paper components.[14] Paper-based packaging contaminated with food is not desirable in paper recycling facilities because of cleaning difficulties, leading to contamination issues.[15]

The quality of paper for recycling largely depends upon the collection system, which is the first step in the recycling process, public environmental awareness, sorting activities, the price for recovered paper and the impacts of printing and converting techniques.[16,17] Besides these factors, the utilization of paper for recycling in the paper industry is affected by its price, processing costs, availability, the production of poorly recyclable products (influence of printing inks and adhesives) and lack of legislation and regulations.[16]

Table 1
CEPI paper and board production and consumption, utilisation of paper for recycling by sector in 2016 [1]

'000 Tonnes Paper grades	Production	Consumption	Grades of paper for recycling				Total use of paper for recycling	Utilisation by sector, %	Utilisation rate, %
			Mixed	Corrugated and kraft	Newspapers and magazines	Other grades			
Newsprint	6.549	5.999	22	0	5.732	131	5.885	12.3	89.9
Other graphic papers	27.360	20.849	129	27	2.986	667	3.809	8.0	13.9
Total graphic papers	33.909	26.849	151	27	8.719	797	9.694	20.3	28.6
Total packaging papers	45.671	39.802	8.351	22.541	492	2.248	33.632	70.4	73.6
Sanitary and household	7.301	7.033	269	126	535	1.882	2.813	5.9	38.5
Other paper and board	4.050	3.716	245	1.044	190	132	1.611	3.4	39.8

Table 2
Composition of paper for recycling

Sample*	% (w/w) Cellulose	% (w/w) Hemicellulose	% (w/w) Lignin	% (w/w) Kaolin/calcium carbonate (ash)	Reference
Copy paper	46	11.9	1	33	[22]
Office paper	84.9	12.3	1.4	1.4	[23]
Coated paper	42.3	9.4	15	-	[24]
Newspaper	48.5	9	23.9	-	[24]
Newspaper	41.02	24.85	23.07	5.99	[25]
Newspaper	68.5	13.1	23.4	3.9	[23]
Cardboard	56.9	10.7	17.8	12.8	[23]

*name given by the authors

The increased recycling rate and the use of commingled collection systems has reduced the quality of the collected paper and of the produced recycled paper.[18,19] An increased use of non-fibre components can lead to increased difficulties in the later stages of the recycling processing chain.[9] During its lifecycle, paper is in contact with different materials, which results in numerous impurities, thus solid particles, moisture and dissolved substances can migrate from one waste material to another, which can have an enormous effect on material recycling.[20,21] For example, the presence of Si, Ti, nutrients and halogens (N, S, Ca, K, Na, P, Cl and F) most likely originates from absorbed food scraps and dust.[21]

Printing inks

Printing inks are coloured complex mixtures, whose main task is to convey a message, provide protection and to give a decorative effect to the substrate to which they are applied. Mostly, they are composed of colorant (pigment or dyes), binder (resins, oils or solvents), solvent (oil- or water-based) and additives (chelating agents, antioxidants, surfactants, biocides *etc*.).[14] Pigments are dispersed solids, insoluble in the support material and almost exclusively used in printing processes. Compared to pigments, dyes are soluble and have lower lightfastness and lower resistance to water. Resins used in printing inks contribute to the properties of hardness, gloss, adhesion and flexibility in an ink.[14] Mostly, resins or polymers are used as binders (phenolic resins, alkyd resins or synthetic resins (polyacrylates)). The ink binder is responsible for ink properties, such as viscosity, drying properties, and surface energy.[26] Oils in printing inks may be drying, semi-drying or non-drying. Drying vegetable oils are glycerides or triglycerides of fatty acids, varying from those that are completely saturated (no double bonds) to those that contain three or more double bonds. Mineral oil, in various forms, is used as a binder in printing inks or as a solvent or diluent in the manufacture of ink binders. It consists of varying percentages of aromatic, naphthenic and paraffinic hydrocarbons, with a small sulphur content ranging up to 4%.[14] Solvents dissolve the binders and adjust the viscosity of the ink for different printing processes.[27] Additives are used to improve the ink properties and are added to ink formulations in amounts not exceeding 5%. These compounds include surfactants to reduce the surface tension and thereby the wetting problem. Adhesion promoters or wetting agents enhance the binding to the printing stock and biocides prevent microbiological degradation of the ink.[27] The main function of a plasticizer (epoxidized compounds, phthalates, polyesters, benzoates) is to make the dried ink flexible and driers help with the oxidative drying process, as well as to supply elasticity to an ink film. Driers are catalysts used to promote oxidation of the drying oils (inorganic salts and metallic soaps of organic acids). After application of printing ink onto a substrate, the binder dries and binds the colorant to the substrate under the press running conditions.[14] Ink drying can be achieved by physical and chemical means or a combination of both, depending on the chemical composition of printing inks.[14] Printing inks are formulated for individual printing processes, classic (offset, flexographic, screen, gravure) and digital (electrophotography, ink jet, laser). They must have certain rheological properties in order to be transferred to the printing plate and afterwards from the printing plate to the substrate (paper).

Paper–ink interactions

The adhesion of inks to paper can be affected by ink formulation, *i.e.* colorants, solvents and vehicles.[14] The influence of colorants on ink adhesion mostly depends upon their compatibility with the vehicle system, their type, percentage and degree of dispersion in the final ink. Adhesion is influenced by the degree of vehicle penetration on paper, while for non-absorbent substrates, it is controlled by the film-forming ability of the resin and the molecular affinity for the substrate. Solvents can affect the adhesion of printing ink in two different ways: enchaining wettability and increasing penetration. An improvement of ink penetration into the substrate can assist physical and chemical bonding. Adhesion can also be affected by adding additives in small amounts during the manufacturing process in order to improve chemical bonding between the printing substrate and the ink.[14]

Interactions between cellulose and ink can be described as physical, purely adhesive or a mixture of both.[28]

Borch[29] and Forsström *et al*.[30] explained that the mechanisms controlling the adhesion between inks and printing substrates are based on:

- Thermodynamic characteristics: surface energy has a central role, as the wetting of cellulose by the ink is important for the final ink–cellulose interaction. Wetting is controlled by dispersion and polar interactions between paper and ink.
- Molecular contact area – surface topography (paper roughness). In electrophotography and thermal printing, a decrease in toner adhesion occurs for rougher papers and similar behaviour is noted for polymer coatings at low coating levels. The contact area governs interdiffusion and entanglement between the constituents of the ink with the amorphous part of the cellulose.
- Fusing temperature, time and pressure.

Surface interactions between printing ink and cellulose are of importance during printing, as well as during ink removal in paper recycling as a high adhesion between ink and cellulose might cause problems during ink detachment.[30] Surface energy studies can explain deinking effectiveness and the fact that different compositions of printing inks may cause different problems due to differences in surface chemistry and mechanical dispersion properties.[31] For example, Vukoje *et al.*[32] explained the poor deinkability of thermochromic offset prints due to creation of very strong bonds, resulting in high adhesion. When it comes to adsorption deinking, the surface free energy of inks, paper and polymers used as adsorbents, the structure and specific surface area of adsorbents can explain the ink detachment and recycling effectiveness.[33]

According to Nie *et al.*,[34] different printing processes will result in the formation of different bonds between inks and paper. During printing, inks undergo different physical and chemical surface property changes. Mineral oil-based offset ink is simply absorbed by the pores in the paper, while water-based ink is physically dried (evaporated) at a temperature below 60 °C. Because no chemical reaction happens during the printing, oil-based offset-heat ink and dried water-based ink keep their original characteristics (hydrophobic/hydrophilic) after the printing process.[34] Vegetable oil is commonly used in offset printing. Over time, the oil components on the fibre surface react slowly with oxygen to form a high molecular weight three-dimensional polymer network with the ink pigment, resulting in the formation of a solid film.[35] In addition, covalent bonds between ink and cellulose are formed.[36]

The photocopy and laser-print toner are made of carbon black, thermoplastic resins and electric–magnetic iron oxide. The thermoplastic resins commonly used are polystyrene, the copolymerization of ethylene and vinyl acetate, nitro cellulose, polyvinyl chloride (PVC), polyamide (PA) and polyester, *etc*.[37,38] Toners polymerize onto the paper surface using thermoplastic binders during a high-temperature printing process.[39] Due to exposure to heat (up to 200 °C), light and oxygen (air), the toner particles undergo polymerization and oxidation, with subsequent formation of peroxide bonds. The polymerization of toners during the printing process results in the formation of chemical bonding between the cellulose fibre and the ink particle and/or physical entrapment of the cellulose fibre within the ink particle.[34] The resins in the toner are melted together with black carbon, fixed by thermal/photothermal fusion, and placed electrostatically on the paper surface in the printing process.[34,37,38]

PAPER FOR RECYCLING AND WASTE MANAGEMENT METHODS

Reducing the amount of all kinds of produced waste streams has been targeted through European laws, either by prevention, reuse or recycling.[40,41] In order to improve the recovery of waste, it should be collected separately, if technically, environmentally and economically practicable, and it should not be mixed with other waste or materials with different properties. In addition, member states should take measures to promote high quality recycling and set up separate collections of waste. The collection of paper for recycling is very high in Europe, according to CEPI it was 56.406 million tonnes in 2016.[1]

Different waste management options for paper for recycling have been studied worldwide, due to different issues occurring. For example, poor deinkability of prints, contamination issues related to food, high moisture content of collected paper, *etc*. Despite the fact that a large percentage of the paper for recycling is used for producing recycled paper and board, different authors put forward various proposals, from incineration, production of bioethanol to biodegradation in aerobic and anaerobic environment. According to European laws, waste management should be followed as presented in Figure 1. The most important is to prevent and to reduce the amount of all kinds of waste.[41] However, if the waste is produced, then it should be recycled (by means of material or organic recycling) or used for energy production.

Material recycling

The recovery of paper for recycling from municipal solid waste is useful, since it can be used for production of different valuable products. The production of recycled paper uses less energy (28-70% energy savings), reduces carbon dioxide emission, the volume and loading of effluent, compared to paper produced from virgin fibres.[17] Paper for recycling and paper industrial residues containing high cellulose content have been also proved to be a promising source of low-cost raw material for the production of different cellulose derivatives, as well as for other high value-added bioconversion processes,[43] such as ethanol[25,44–49] and methane production.[23,50] Pendyala *et al*.,[51] Argun and Onaran,[52] Eker and Sarp[53] have studied the potential of paper for recycling for hydrogen production, but they did not describe the influence of prints on hydrogen yield.

Production of recycled fibres by deinking

Deinking is the most important process in the production of recycled paper and depends upon the quality of the collected paper for recycling, the type of printing process and the properties of the printing inks, the age of the product and climatic conditions during its life cycle.[34,54–56] In order to achieve desirable properties of recycled pulp (optical and mechanical), improved deinkability of printed paper products has become an essential factor for a potential use of recovered paper in graphic papers production. Moreover, deinkability of packaging grades is also of great importance, due to the growing tendency to print certain products, such as cardboard.[57] When colloidal printing ink dissolves in the water and colours the entire pulp, a serious problem occurs in the case of graphic paper production, but it is not significant for brown paper production.[58] Packaging paper grades and mixed grades are commonly recycled without deinking and the main task is to disintegrate the packaging board into a suspension. The deinking process is common practice for graphic paper grades, except some high-quality grades with little or no ink.[13] Recently, the amount of paper-based packaging products exceeded that of graphic products in the collection system. Mostly, corrugated boxes and folding boxboards have the largest share among paper-based product categories and are normally quite easy to recycle in standard paper mills.[59] Researchers suggest that brown packaging, corrugated and paper boards should be collected separately and recycled separately from graphic paper grades due to unsatisfying recycled pulp properties. Iosip *et al.*[60] showed that packaging paper and board from household collection, strongly affects the optical properties of deinked pulp, by decreasing brightness and by increasing the number and size of specks due to brown fibre flakes.

Different deinking processes can be used for the production of recycled fibres, based on the quality of paper for recycling, as well as on the desired quality of the produced pulp and the requirements of the pulping process, such as the chemical or enzymatic approach. The most common methods for ink removal are based on the chemical approach, thus the proper selection of deinking chemicals is often a compromise between costs and performance.[61,62]

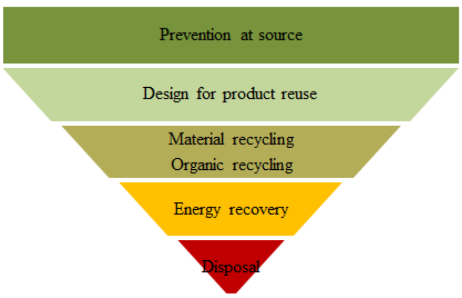

Figure 1: Waste management hierarchy[42]

Detachment of ink particles during deinking

Deinking involves ink detachment from the fibre and ink separation from the pulp suspension. Ink detachment occurs in the pulper where chemical and mechanical actions release ink from the fibre, while ink separation occurs in the flotation and washing stages.[35,63] The separation method is based on particle surface chemistry and system hydrodynamics. The theory of removing ink from paper fibres by flotation is chemistry based; however, there are some key physical aspects involved (bubble population and size, degree of turbulence in the suspension). As the ink particles collide with bubbles, bubble–ink aggregates are created. The ink-laden bubbles can then be removed, provided that appropriate hydrodynamic conditions are available within the flotation cell.[63]

According to Borchardt,[64] the ink detachment mechanism can be divided into four steps:

1. Surfactant-promoted solubilisation into the aqueous pulping medium;
2. Surfactant-promoted wettability alteration of cellulose surfaces, promoting ink detachment and emulsification;
3. Cellulose fibre swelling, which reduces ink adhesion to the fibre; fibre swelling is promoted by high pH;
4. Cellulose fibre bending and inter-fibre abrasion promoted by mechanical agitation.

Ink detachment as a result of fibre swelling is assumed to be due to the inability of ink to swell to the same extent as the fibres.[35]

According to Stack *et al.*,[65] most of the ink is believed to be held onto the fibre surface by weak van der Waals forces, which can be detached through mechanical shear and interfibre abrasion in the pulper. Ink in direct contact with the fibre surface is believed to be held by hydrogen bonding. The use of chemicals (alkali and surfactants) is needed to detach these ink particles.[65] Alkali is added to swell the fibres and soften the ink through saponification. Surfactants can alter the wetting ability of the fibres, to improve alkali penetration and also to help keep the detached ink dispersed and prevent it from re-depositing.[65] Dispersants prevent re-agglomeration and re-deposition of ink onto the fibres, so that the ink can be removed during a washing or thickening stage.[28] Calcium soaps of fatty acids are the most commonly used surfactant system in the flotation deinking process, acting as collectors and enhancing ink removal by precipitating as particles on the ink particles. Following this process, the ink particles become hydrophobic, agglomerate and attach more efficiently to the air bubbles.[28]

In enzymatic deinking, enzymes can act directly either on the fibres or on the ink film.[39,62,66] Cellulases and hemicellulases remove inks from recycled paper by peeling off the fines and small fibrils on fibre surfaces.[67] When cellulases/hemicellulases are used, the release of ink particles into the suspension is generally attributed to the cellulose hydrolysis on the fibre/ink inter-bonding regions, which facilitates ink detachment. These enzymes can remove small fibrils from the surface of the ink particles, thus altering the relative hydrophobicity of the particles, which facilitates their separation in the flotation/washing step.[39,62,66] Laccases are multicopper oxidases that catalyse the oxidation of phenolic compounds and aromatic amines using oxygen as electron acceptor.[68] They can remove the surface lignin in the presence of some low molecular mass mediators and oxygen by oxidation of lignin moieties, oxidative cleavage of side chains and selective oxidation of free phenoxy groups in lignin.[69] Thus, they have some potential in deinking old newsprint, as they may selectively remove surface lignin to facilitate ink removal.[70] Lipases have shown some direct action on ink particles, either degrading oil carriers or breaking down pigments.[71] Cutinases catalyse the hydrolysis of a variety of soluble esters and insoluble triglycerides, (ester bond) in natural resin, synthetic resin, and polyester, which were used in the ink vehicles. That caused the pigment to fall from the paper surface and be removed with air bubbles.[71]

Deinking by flotation

Currently, industrial processes related to paper recycling are mostly dealing with washing and flotation. Washing is most effective for removing small particles (<10 µm), while flotation – for medium-sized particles (10-150 µm). Screening and centrifugal cleaners are used for removing large ink particles (>100 µm).[62] Washing is used for the removal of particles smaller than the mesh size of a filter cloth, mostly of fillers and coating particles, fines, microstickies and ink.[58]

Deinking by flotation is the most common practice for ink removal in the paper recycling process, in which hydrophobic ink particles are removed from the pulp suspension by adhesion to air bubbles, forming a froth, which can be separated from the pulp suspension. The efficiency of the deinking process depends on several factors, such as fibre and ink properties (pigment size and type of solvent used), printing conditions, bubble size, collectors and calcium soaps formed from calcium ions and fatty acids, and paper surface properties.[72,73] For example, inks printed on coated paper surfaces detach more easily than inks printed directly on uncoated paper surfaces.[73]

The paper mills that produce recycled paper or cardboard have abundant cleaning systems (sorting machines and for graphic papers deinking systems). After defibrating, the suspension passes through several successive cleaning systems in which impurities are separated by their density, size or shape.[20] Recycling plants can mostly handle a certain amount of unwanted non-paper components, but if the amount gets too high, the recycling process can become less economical. However, paper for recycling, which usually comes with different types of applied prints, will not always show positive deinkability results (Table 3). Deinking by flotation is efficient mostly in the case of conventional offset and gravure printing inks, where hydrophobic printing inks are present.[54,58] According to Faul,[13] 81% of the offset prints, mostly newspapers and magazines, achieved a positive assessment of their deinkability, while the latter often occurs in the case of UV-cured prints. Digital printing inks or water-soluble inks cannot be deinked, or only with great difficulty. Toner prints are usually well deinkable, if made by a dry toner process, but in the case of liquid toner prints the test fails, because of the very high content of dirt particles in the deinked pulp.[74] Water-based flexographic inks present a particular problem to the deinking process because of their small particle size (0.2-1.0 µm) after re-pulping, and they are too small for removal by flotation. In addition, these inks are hydrophilic and cannot be agglomerated. For the successful removal of flexographic inks, recycling processes therefore incorporate aggressive washing stages, which can lead to economically unacceptable yield losses.[31,75]

Also, deinkability problems can be noticed in the case of UV-curable coating because of visible speck contamination and of water-based dye prints because of low brightness or/and pronounced colour shade.[76] During digital printing (photocopy and laser-printing), toner particles are exposed to heat (200 ºC), light and oxygen, which results in the ink fusing with the paper and making it non-dispersible.[75,77] During printing, the ink particles undergo the process of polymerization and oxidation with subsequent formation of peroxide bonds, resulting in strong chemical and physical bonding with cellulose fibres. During flotation deinking, these thermally fused ink particles and cellulose fibres will lead to formation of larger ink particle sizes, resulting in poor deinkability. Besides polymerization, oxidation will create a greater polarity at the toner particle surface, which can reduce the deinking efficiency.[34] Balea *et al*.[78] showed that an improvement in the removal of water-based inks can be achieved by addition of cellulose nanofibres in combination with a cationic polyacrylamide.

Deinking efficiency is also affected by the influence of light, temperature, moisture and storage time.[55,56] The age of printed products mainly affects offset prints.[13] Pan and Nguyen[79] showed that in the early stage after printing of offset ink, it is more difficult to detach mineral oil based ink than vegetable oil based ink, but after thermal ageing, the energy required for ink detachment is similar for both types of ink.

In addition to conventional printing inks that have been used so far, the development of new materials has increased and can affect the efficiency of recycling, regardless of the printing process, which they have been made for. Small variations in their chemistry can affect their quality, printing quality, as well their deinkability. As the ink formulation is an essential factor in deinking, it is crucial to examine deinkability aspects of novel printing inks, which may differ from conventional inks in formulation and size of colorants or additives. For example, information about the recycling of functional and smart printing inks is very limited. Aliaga *et al.*[80] reported about the effects of printed electronics on paper recyclability. The analysis was based on a case study focused on envelopes for postal and courier services provided with these intelligent systems. According to the results of the pilot scale recycling tests, the resistors (silicon-based electronic component) do not disintegrate in the pulping stage and they are retained in the screens and thus do not affect the quality of recycled paper.[80]

Table 3
Deinkability of prints

Prints	Deinkability	Nature of ink particles	Problems	Reference
Offset	Good	Hydrophobic	After aging, bad ink detachment	[13, 54]
Gravure	Good	Hydrophobic	-	[13]
Flexographic	Poor	Hydrophilic	Small size and hydrophilic nature of the ink particles which cannot attach to the collectors in the flotation process	[13, 58]
Digital	Poor	Hydrophilic	Generating numerous ink particles above 100 µm	[54]
Inkjet	Poor	Hydrophilic	Ink may stain the fibre, formation of small particles	[31, 81, 82]
Hot melt based ink jet prints	Poor	Fused during drying – residual toner	Sticky deposits	[76]
Toner	Poor	Fused during printing	Formation of larger particles, flat and plate like particles	[31, 34]
Liquid toner	Poor	Too soft to pass the screens	Large visible inked film specks	[76]
UV curable	Poor	Formation of cross-linked films which are difficult to break down	Visible speck contamination by large flat and plate-like particles	[31]

Table 4
The use of enzymes in deinking processes

Prints/paper substrate*	Enzyme	Reference
Mixed office wastepaper	Enzyme extract (endoxylanase and endoglucanase) from fungi *Aspergillus terreus* and *Trichoderma viride*	89
Mixed office wastepaper (laser, inkjet and photocopy prints, photocopy paper)	Commercial endoglucanase, cellulase and xylanase	66
Inkjet-printed paper	Isolate from *Vibrio alginolyticus*	77
Mixed office wastepaper	Commercial endoglucanase, cellulase and xylanase	100
Laser printed office waste papers	Cellulase and hemicellulase from *Aspergillus niger*	91
Fresh and aged recycled newsprint/magazines	Cellulase	67
Old newsprint	Commercial hemicellulase with laccase-mediator system	69
Laser printed paper	Bacterial xylanase from *Bacillus sp.*	39
School waste paper	Xylano-pectinolytic enzymes from *Bacillus pumilus*	84
Old newsprint, magazines, laser, inkjet and Xerox	Bacterial laccase and xylanase from *Bacillus halodurans*	90
Laser printed paper; old newspaper (flexographic); photocopy paper (photocopy dry toner); glossy magazine paper; bubble jet-printed waste paper	Commercial cellulase and hemicellulase	61
Photocopier waste papers (Xerox black toner)	Cellulase and xylanase of newly isolated fungal strain *Trichoderma harzianum*	88
Flexographic inks	Fungal laccases from *Coriolopsis rigida* and *Myceliophthora thermophila*	68
Laser printed paper	Lignocellulolytic enzyme from fungus *Penicillium rolfsii*	38
Old newsprint	Cutinases from *Thermobifida fusca* and *Fusarium solani pisi*	71

*name given by the author

The results showed that nanosilver inks were dissolved in the pulp suspension and can affect both mechanical and optical properties of the recycled paper. In addition, the authors have emphasized that real impacts on industrial recycling are expected to be even significantly lower, since the proportion of paper products with printed circuits in the current paper waste streams are much lower. However, they also point to the fact that further research on the treatment of printed electronics and smart labels is needed, in parallel with innovation in these communicative devices.[80] The study of thermochromic offset printing inks showed they are very difficult to deink by the flotation process, which is a successful method for conventional offset printing inks.[32] Some problems in recycling can be avoided by using enzymes or new methods for ink removal, such as ultrasound and adsorption deinking.

Enzymatic deinking

Enzymatic deinking is a valuable approach due to its high efficiency and environmentally friendly nature.[83] Enzymatic treatment can improve the physical properties of handsheets, while decreasing pollution of effluents, thus contributing to the reduction of environmental pollution and lowering wastewater treatment costs.[62,84] According to Singh *et al.*,[84] the use of chemicals can be reduced to 50% to attain the same extent of brightness of handsheets, by combining enzymatic deinking with conventional deinking.

Cellulase, hemicellulase (*e.g.* xylanase), pectinase, lipase, laccase *etc.*, are enzymes employed in the deinking processes. Hemicellulase and cellulase can attack the components on the fibre surface. Detachment of ink from fibres takes place when these enzymes alter the fibre surface by modifying chemical bonds. Hemicellulases can result in the hydrolysis of hemicelluloses; starch based coating can be hydrolysed by amylolytic enzymes, lipase can degrade vegetable oil based ink binders and lignin is removed by the laccases.[62,85] The use of different enzymes reported in the literature is presented in Table 4. Depending on the used printing ink, printing process and paper type, the proper enzyme in the enzymatic deinking process should be chosen. By the use of enzymatic deinking, the increase of the recycled pulp optical properties (brightness, whiteness, ERIC), reduction of ink particle size, improved drainage and better mechanical properties can be achieved.[61,62,86,87] It works in neutral or slightly alkaline environments, which reduces overall chemical requirements and minimises yellowing of reclaimed paper after alkaline deinking. Enzymatic deinking is most effective when carried out in the presence of surfactants, which increase the performance and stability of the enzyme.[84] Enzymatic technology and cellulolytic enzymes have shown the most promising results for deinking of mixed office waste (MOW).[66,67] MOW reuse is usually limited by the high content of toners, which are very difficult to remove by the conventional flotation deinking because of the thermoplastic binders that polymerise and fuse onto the paper fibres during the high-temperature printing process, resulting in strong adherence of toner ink particles to fibres.[66,83] When these fibres are chemically treated, the toner particles usually remain as large, flat, rigid particles that separate very poorly from fibres during the fibre/ink separation stages.[66] These problems can be overcome by the use of enzymes. Enzymatic deinking in the case of flexographic inks can be influenced by the presence of paper additives, and the highest inhibition was reported for calcium carbonate.[68]

Enzymatic deinking may improve the strength properties of paper by removing the fines content and improving the interfibrillar bonding of paper.[61] Pathak *et al.*[87] reported the improvement of burst and tensile indices, by 15.3% and 2.7%, respectively, the decrease of brightness and tear index by 2.1% and 21.9%, respectively, by deinking photocopy paper using commercial cellulase enzyme. Lee *et al.*[61] showed that commercially available enzymes, cellulase and hemicellulase, increased the tensile index of magazine paper, but reduced the tensile index of bubble jet-printed paper, photocopy paper and newspaper. Enzymatic hydrolysis caused a 21.1% reduction in the tear index for bubble jet-printed paper, but a 3.1% increase in the tear index was obtained for laser-printed paper, relative to the blank.[61] In addition, enzymatic hydrolysis increased the burst index by 4.7%, relative to the blank, for laser-printed paper. For the photocopy paper, the highest reduction (8.3%) in the burst index relative to blank was noticed.[61] Crude cellulase and xylanase isolated from the fungal strain *Trichoderma harzianum* used for deinking of photocopier waste papers showed 23.6% higher deinking efficiency and 3.2% higher brightness of deinked pulp with respect to chemically deinked pulp.[88] In addition, the increase of pulp freeness (21.6%) tensile index (6.7%), burst index (13.4%) and folding endurance (10.3%) of handsheets was observed. However, tear index was decreased by 10.5%.[88] Marques *et al.*[89] showed that enzymes extracted from xylan-grown *Aspergillus terreus* and cellulose-grown

Trichoderma viride contributed to the improvement of the strength properties, relative to the control, in the three strength tests (tensile, burst and tear), with the exception of the tear index, which was not affected by *A. terreus*.[89] For the enzymatic deinking of wastepaper (old newsprint, magazines, laser, inkjet and Xerox) by laccase and xylanase, increases in brightness (21.6%), breaking length (16.5%), burst index (4.2%) tear index (6.9%), viscosity (13%) and cellulose crystallinity (10.3%), along with a decrease in kappa number (22%) and chemical consumption (50%), were observed by Virk *et al.*[90]

Even though most of the conducted studies show an increase in the physical properties of recycled pulp after enzymatic treatment, ligno-cellulolytic crude enzyme from *Penicillium rolfsii* was found to reduce the paper strength properties, based on the results of tensile, tear and burst indexes, most probably due to cellulose degradation.[38] Decreases of 32% in tensile strength, 60% in tear index and of about 52% in burst index were noticed after enzymatic deinking of laser-printed paper.[38] According to Lee *et al.*,[91] lower tensile strength in the enzymatically deinked papers is expected due to the breakage of the fibres, while the degree of fibre breakage depends on the extent of the synergistic effect of the enzymes.[91]

According to Bajpai,[62] enzymes for deinking are now commercially available and at lower cost than in the past, while their increased use and the advances in fermentation technology are expected to lower the production costs of enzymes, enabling their use in mill-scale processes.

Adsorption deinking

In adsorption deinking, the ink particles are adsorbed onto the surface of polymer blends (adsorbents), instead of air bubbles (in deinking flotation), at high stock consistencies of 15% and, at the same time, have a dispersing effect (ball mill effect); 90% less water needs to be pumped in the facility owing to the high stock consistency.[58] The efficiency of this method can be affected by the concentration of calcium ions, the deinking chemicals used, the polymer blends used as adsorbents, the printing ink and paper used.[92] Besides these factors, the efficiency of the method is affected by the surface free energy of inks, paper and polymers used, their structure and specific surface area.[33] Researchers have studied the possibility of using different polymers in adsorption deinking. Darwish *et al.*[93] showed that polyethylene (PE) has a certain capability of removing toner particles, while the wax-coated PE enhances the removal of toner particles beyond that obtained using only PE.[93] Liu *et al.*[94] investigated the use of PET films in deinking of xerography wastepaper and the obtained results showed that, by choosing the proper combinations of solvent, surfactant and toner, viscous liquids can be formed, which can be adsorbed onto plastic films at ambient temperature and separated by a screen process. Du *et al.*[82] studied the potential of using chitosan as an adsorbent for removal of ink jet printing inks, which showed an increase of ISO brightness compared to standard deinking flotation procedure INGEDE method 11. Petzold and Schwarz[95] showed that the efficiency of adsorption deinking is strongly influenced by the properties of suspensions, which depend on the type of paper. Jamnicki *et al.*[96] showed that adsorption deinking is more successful in the reduction of mineral oils in recycled pulp than the conventional flotation deinking method (INGEDE Method 11). The main conclusion was that over 60% and up to 80% of mineral oils can be removed from paper for recycling by means of adsorption deinking.[96]

Ultrasound deinking

The studies dealing with the ultrasonic deinking of paper for recycling are mostly related to the removal of digital prints, which are not easily removed by conventional flotation methods. Ultrasound deinking uses cavitation to separate the printing ink particles from the fibres in high stock consistencies.[58] The cavitation occurs at frequencies between 20 and 100 kHz, when ultrasound energy turns liquid into vapour at nucleating sites within the liquid.[97] The produced bubbles grow in size before collapsing and releasing energy (implosion).[97] In the paper processing industry, ultrasound can be used at different stages: enhancement of pulping, bleaching, depolymerisation of cellulose and treatment of wastepaper.[98] The main advantage of using sonication resides in the fact that experiments can be carried out at ambient temperature under atmospheric pressure, contrary to other advanced oxidation processes.[98] Tatsumi *et al.*[99] also concluded that ultrasonic treatment can be effective for offset-printed newspaper and laser-printed office paper on commercially available paper for xerography, followed by conventional flotation. The process, which consists of ultrasonic treatment for 1 min, following flotation deinking, requires about 1.4 times as much energy as the conventional flotation deinking process, but it induced 20% improvement in brightness.[99]

Manning and Thompson[97] have used high intensity ultrasound for removal of UV curing screen printing ink. Exposure of pulps to high intensity ultrasound causes breakdown of the ink film under neutral pH conditions and detachment of significant proportion of UV cured ink particles from the pulp slurry.[97] According to Fricker *et al.*,[101] the ultrasound is effective in removing indigo inks as well. It can remove almost 100% of ink from pulp suspensions because it causes the breakdown of ink particles to floatable sizes, which can be successfully removed by flotation deinking.[101] The influence of temperature on an ultrasound-assisted ink removal process has been investigated by the Gaquere-Parker *et al.*,[98] who showed the effectiveness of ultrasound at any temperature tested (15-45 ºC), but brightness data and UV-Vis absorbance indicated the optimum temperature was between 30-35 ºC.[98]

Toxity of effluents from paper recycling processes

Recycling paper mills produce effluents with different key pollutants from those of paper mills using virgin fibres (wood, agricultural residues). The processes of pulping and bleaching are the major sources of pollutants in paper industry effluents.[102] The type and quality of the raw material (wastepaper) used for recycling, as well as pulping and deinking additives, mineral oils and other substances, will affect the quality of the effluents from paper recycling mills.[102]

Table 5 shows that different deinking processes (conducted at laboratory scale) and raw materials used (paper and prints obtained) will have different values of organic components in effluents. Current deinking processes depend upon the use of a large amount of chemicals that produce toxic effluents with high chemical oxygen demand (COD) values.[84] In general, the most commonly used alkaline systems generate heavily loaded process water and thus neutral deinking could be a better solution.[54] Sometimes, surfactants applied in neutral deinking may contribute to the increase of chemical oxygen demand (BOD) and COD values.[103] Enzyme deinking generally decreases the BOD and COD values of effluents, thus lowers the effluents treatment cost and environmental pollution.[62,83,84,86,88] However, in some cases, the COD release appeared to be higher with the enzymatic treatments. This behaviour was described by Magnin *et al.*[85] during enzymatic deinking of mixed wood-free paper (offset, laser and copy) due to the hydrolytic property of the enzymes, since they release soluble sugars from the pulp to the process water, resulting in higher COD. The COD of the pulp slurry effluent of the offset print is lower than that of the industrial newsprint/magazine furnish, since the industrial raw material contains more soluble and colloidal contaminants than the offset print.[104] There is lack of data describing the influence of other methods (ultrasound or absorption deinking) on effluent quality.

Since different studies show different values obtained for COD and BOD values, in order to get more reliable data, the index of biodegradability (IB) should always be calculated according to Equation 1:[88,103]

$$IB = \frac{BOD}{COD} \cdot 100\ [\%] \qquad (1)$$

where IB – biodegradability, %; BOD – biochemical oxygen demand, COD – chemical oxygen demand.

Due to very high concentrations of organic compounds and the presence of toxic compounds resistant to biodegradation, the IB index in paperboard recycling plant effluents is usually below 0.15, which suggests that these effluents are resistant to biological treatment.[105] According to Birjandi *et al.*,[106] samples with the IB smaller than 0.3 are not appropriate for biological degradation, as for complete biodegradation the effluent must present an index of at least 0.40. This means that biological treatments, as stand-alone treatments, are usually ineffective methods for treatment of such industrial effluents.[105]

When it comes to paper recycling plants, studies show that effluents contain complex matrices with a variety of toxic compounds.[107,108] In addition, recycling paper mills generate effluents that contain high concentrations of organic compounds, such as lignin, cellulose and resins.[105] The quality of synthetic effluents derived from laboratory paper recycling, which contains only a limited number of toxic organic compounds are entirely different from real pulp and paper effluent chemistry and behaviour, because of the combination of various chemicals, suspended particles, nutrients and bacteria.[108] In addition, Table 6 shows the physical-chemical characterization of real effluents from paper recycling mills. COD and BOD values are significantly higher than the values obtained for laboratory deinking processes (Table 5). According to Muhamad *et al.*,[108] the six most important wastewater quality parameters, namely, COD, turbidity, ammonia (expressed as NH_3-N), phosphorus (expressed as PO_4^{3}-P), colour and suspended solids (SS), should always be monitored. Besides these parameters, authors also found the presence of other compounds in effluents, such as sulfate,[109] AOX, metal ions, such as Al, Fe, Ca,[110] Si and Na.[111]

Different studies showed the presence of a wide range of various organic contaminants in effluents from paper recycling factories, receiving rivers and sediments, such as Bisphenol A (BPA),[102,107,112] 2,4,7,9-tetramethyl-5-decyne-4,7-diol (TMDD),[102,113] aromatic sensitizers,[114] aryl hydrocarbons including chlorinated aryl ether.[115] Some of those constituents are related to the paper and graphic industry, such as photoinitiators, ink and thermal paper constituents. For example, TMDD is a non-ionic surfactant used in printing inks. Concentrations of TMDD were found to be present in wastewater from factories processing recycled paper from 113 μg/L[113] up to 1700 g/L.[102] The presence of BPA was found to be around 3400 g/L, respectively.[102] The isolated aryl hydrocarbons and aryl ether, as well as aromatic sensitizers, are chemicals used in thermal papers.[114,115] Some of these compounds may affect vertebrate physiological and reproductive functions.[116] For example, adsorbable organic halides (AOX) may bioaccumulate in fish tissue, causing a variety of carcinogenic, endocrine, clastogenic and mutagenic effects, which may then pose problems to humans consuming the contaminated fish.[108] TMDD can be harmful to fish and aquatic invertebrates only at high concentrations.[113] Resin acids and, to a smaller extent, the unsaturated fatty acids can be toxic to fish.[107] BPA and some surfactants commonly present in paper recycling mills are considered as endocrine-disrupting compounds, which, at low concentrations, may potentially alter the normal hormone function and physiological status of animals.[107]

Because of the very high concentrations of recalcitrant organic compounds and resins, the effluents must be treated before being discharged into the environment.[105] Thus, the effluents should satisfy compliance levels, such as chemical oxygen demand (COD), biochemical oxygen demand (BOD), suspended solids (SS) and colour and residual organic matter consisting of potentially toxic chlorinated compounds.[108] Sometimes, the effluents contain soluble organics and particulate matter that are not effectively degraded by traditional wastewater treatment technologies.[117] In order to achieve desirable parameters, different treatment processes have been proved to be efficient and appropriate for reducing organic loads and enhancing biodegradability. Some examples are aerobic sequencing batch reactors with granular activated carbon,[108,118] bench scale horizontal-flow anaerobic immobilized biomass (HAIB) reactor,[119] coagulation process,[120] ultrafiltration,[121] electrochemical peroxidation process.[105] In addition, if the paper recycling mill involves recirculating waters, the presence of organic components can have an impact on the paper machine and product quality.[107]

Health and safety issues related to paper recycling

Recycling is supported for the sustainable use of materials, but on the basis of present toxicological assessments, it is often far beyond acceptable.[122] Pivnenko *et al.*[5] showed that the quality of the collected paper for recycling may ultimately decrease as more and more "marginal" paper fractions are collected for recycling and the contents of harmful substances in paper thereby increase. In addition, they showed that paper for recycling might potentially contain a large number of hazardous chemical substances, while many of them can be associated with the printing industry.[5] If the printing inks are not entirely removed during paper recycling, the recycled pulp and the product made from it may contain residues.[5,123] Paper and cardboard, partly or fully produced from recycled fibres, can be used in contact with dry foodstuff such as flour, grain, sugar, salt, rice and pasta, and as such, must meet a certain basic set of criteria concerning safety issues.

Table 5
Organic pollution of effluents from paper recycling effluents generated at laboratory scale by different deinking processes and samples used

Sample	Deinking process	Parameter		Reference
		COD (mg/L)	BOD (mg/L)	
Offset	Chemical – alkaline	2650	-	[54]
Digital		4950	-	
Flexographic		2150	-	
Flexographic	Chemical – neutral	1309	-	[54]
School wastepaper with blue ink	Chemical – alkaline	3750	1136	[84]
	Enzymatic – xylano-pectinolytic	1056	368	
Toner	Chemical – alkaline	270	122	[88]
	Enzymatic – commercial	148	85	
	Enzymatic – isolated	168	99	
Offset	Chemical – alkaline	779	-	[104]
Newspaper/magazine		921	-	
Mixed – flexographic toner and offset	Chemical – alkaline	409	240	[103]
	Chemical – neutral	479	310	

Table 6
Physical-chemical characterization of real wastewater from paper recycling mills

COD (mg/L)	BOD (mg/L)	Sulfate (mg/L)	Ammonia (mg/L)	Phosphate (mg/L)	SS (mg/L)	Turbidity (TNU)	Colour Pt-Co	AOX	Reference
1057	-	-	4.1	0.03	149	73	121	-	[108]
4300	535	-	-	-	-	1308	-	-	[105]
7820	3963	79	-	-	-	-	-	-	[109]
3523	940	50	-	73	260	873	-	-	[120]
1152	-	-	-	-	-	-	-	249	[118]
1500	563	-	0.17	-	2138	-	1002	-	[124]
780-800	150-200	-	170-200	-	-	-	-	-	[125]

Different authors have studied the contaminants in recycled fibres, mineral oil hydrocarbons, phenols, phthalates, polychlorinated biphenyls, bisphenol A and toxic metals.[5,123,126–129] In a study on recycled paper containing thermochromic offset inks, Jamnicki Hanzer *et al.*[130] showed the presence of Bisphenol A originating from thermochromic inks. Vápenka *et al.*[131] showed the presence of 68 contaminants in packaging materials originating from paper pulp processing residues, printing inks or adhesives (photoinitiators, plasticizers, solvents), impregnation and coating (solvents, hydrocarbons). Mertoglu-Elmas[11] showed that coloured pigments are sources of heavy metals (Pb, Cd, Zn and Cu) in recycled paper. Most of the contaminants in paper packaging materials originated from materials used for modification of functional properties and/or residues from recycled pulp.[131] Since recycled cardboard is made by disintegration of different mixtures of materials (newspaper, journals and cardboard) without ink removal, recycled pulp can contain mineral oil saturated hydrocarbons (MOSH) and mineral oil aromatic hydrocarbons (MOAH) mostly originating from printing inks in newspapers.[122] The mineral oil content could be reduced through the selection of the paper and board fed into the recycling process, by replacement of mineral oil containing inks with inks free of mineral oil (flexographic, digital, UV curing inks), which would also enable efficient deinking.[132]

Bioethanol production

Bioethanol derived from lignocellulosic biomass is a valuable renewable energy source, recently used as a substitute for fossil fuels in road transport. Paper for recycling has relatively high carbohydrate content – up to 80%, and a higher amount of cellulose than other lignocellulosic materials, which gives them considerable potential as feedstocks for bioethanol production.[44,48] It has been estimated that an annual yield of 82.9 billion litres of cellulosic bioethanol could be derived from paper for recycling worldwide, replacing 5.36% of gasoline consumption with GHG emissions savings up to 86%.[44] Bioethanol production from paper consists in hydrolysis and fermentation. The steps can be accomplished in two stages (separate hydrolysis and fermentation – SHF) or in a single stage (simultaneous saccharification and fermentation – SSF).[133] An enzymatic hydrolysis of paper for recycling is becoming a perspective way to obtain raw material for the production of liquid biofuels, as presented by different authors. [44,46,49,134–136] Besides paper for recycling, the sludge from pulp and paper production can also be useful for bioethanol production.[133,137–139] The presence of printing inks, fillers and other additives makes enzyme hydrolysis into fermentable sugars difficult, because of ineffective adsorption of the enzyme onto the substrate caused by possible binding of a proportion of the enzymes to impurities in the substrate (ink particles and paper fillers) or binding with lignin.[44,140] According to Wang *et al.*,[44] enzyme activity is affected more by product inhibition (high lignin and glucose content) rather than by ineffective adsorption caused by impurities, such as fillers or inks. Calcium carbonate inhibits cellulase activity remarkably, as presented by Schroeder *et al.*,[133] Min and Ramarao[141] and Wang *et al.*[142] According to Schroeder *et al.*,[133] enzyme activity can also be influenced by aluminium and silicon originating from the fillers used in the papermaking process and metals from the printing inks. Despite the fact that printing inks can inhibit enzyme reactions, and thus decrease the efficiency of bioethanol production, in the presented literature, the influence of printing inks on the efficiency of bioethanol production is not taken into account. Only Guerfali *et al.*[134] have reported the importance of a washing pretreatment process of paper for recycling in order to remove printing inks. Enzyme inhibition problems can be avoided by using different pretreatment methods, such as ethylene glycol pretreatment, ultrasound in fermentation process, delignification alkali (NaOH) and enzymatic (ligninolytic) pretreatments of waste newspaper, or use of accelerants and hydrogen peroxide during the paper mill sludge conversion.[25,138,140,143] Although the literature references mention possible inhibition of enzyme activity by the various substances that may originate from the printing inks, there is no available information on how certain printing inks (*e.g.* offset, flexographic, UV curable, non-impact printing inks) affect the efficiency of bioethanol production from paper for recycling.

Production of cellulose derivatives

Due to the higher cost of recycled paper production and its lower quality, a number of studies have been conducted in order to find an alternative option for utilization of paper for recycling, including cellulose micro- and nanocrystals, cyanoethyl and carboxymethyl cellulose synthesis and biocompatible cellulose aerogels, as presented in Table 7. The quality of the raw material used for the production of cellulose derivatives can remarkably influence the quality of the products and process yields.[144] The presence of residual ink, hemicellulose and lignin in paper for recycling are highly undesirable when it is used for production of cellulose derivatives, and thus these compounds should be removed.[6] According to the literature presented in Table 7, pretreatment methods and deinking by flotation may ensure the removal of undesirable amorphous-type polymer components.[145] Moreover, alkali treatment can disrupt hydrogen bonding in the network structure between different cellulose chains, improving the effectiveness of subsequent treatments, such as the acid hydrolysis process.[6,146] With the inclusion of a pretreatment process before acid hydrolysis, an increment in quality can be achieved, favouring the access of the acid to cellulose.[144]

Table 7
Production of cellulose derivatives from paper for recycling

Product	Source*	Pretreatment	Reference
Rod-like cellulose nanofibres	Waste newspaper	Alkaline treatment and bleaching	147
Cellulose nanocrystals	Recycled pulp (1% lignin) produced from waste commercial wood pulp and used business papers	-	148
Cellulose nanocrystals	Waste paper	Alkali and bleaching treatments	149
Cellulose nanocrystals	Old corrugated container fibre	Mechanical (pulping) and alkali pretreatment	150
Nanocrystalline cellulose (NCC)	Non-printed areas of recycled newspaper	Alkaline treatment and bleaching	151
Cellulose nanocrystals	Office waste paper (OWP)	Deinking and defibering by use of chemical agents and mechanical force, performed by washing through a fine mesh	4
Cellulose nanostructures	Primary paper residue	Mercerization followed by bleaching	43
Cellulose nanocrystals	Old newspapers and recycled newsprints	With and without alkali and bleaching treatments	144
Microcrystalline cellulose (MCC) powder	From three grades of waste paper: book, ground-wood/newsprint and paperboard	Deinking	152
Cellulosic aerogel from cellulose nanocrystals	Old newspaper	Alkali and bleaching treatments	145
Cellulose-based films	Waste newspapers	Alkali treatment and bleaching	153
Cyanoethyl cellulose	Office waste paper OWP (photocopier and computer print-out papers)	Deinking by flotation	6
Carboxymethyl cellulose	Mixed office waste (MOW) photocopier and computer print-out papers	Pulping and deinking by flotation	7
Highly porous cellulose beads	Printed paper wastes	Pretreatment and maceration processes in alkali solution	154
Cello-oligomers	Printed paper wastes	Pulping and alkali treatment	155

*name given by the author

The main drawback of the pretreatment methods is their high chemical and energy consumption, resulting in cost increases, and the low process yield because of the high pretreatment losses.[144] Paper for recycling is an attractive potential feedstock for production of valuable cellulose derivatives and its utilization could decrease the demand of competitive and conventional raw materials, such as cotton fibres and wood pulp for modified cellulosic products.[7] Despite the fact that authors emphasize the importance of printing inks removal from the stream of paper for recycling, in order to get higher efficiency in the production of cellulose derivatives, there is lack of information in the presented literature regarding ink removal during conducted pretreatment methods and deinking. Also, most of known studies are dealing with mixed office paper for recycling printed with non-impact printing technologies, which is generally known to be difficult to deink by conventional deinking by flotation. In addition, there is no available information about the influence of other printing inks on the efficiency of cellulose derivatives production. Due to different chemical formulations of printing inks, we suggest that, in the future, some other printing inks and printing techniques should be also taken into account when investigating the production of different cellulose derivatives from paper for recycling.

Organic recycling

Even though most collected paper for recycling will generally be recycled by means of recycled paper production, some of it will be contaminated with food or wet (about 40%). In these forms, it is not suitable for conventional recycling methods, but is suitable for organic recycling.[8,156] Organic recycling is also considered as material recycling due to production of compost or biogas. However, the main problem is that, as with incineration and bioethanol production, when paper is composted or digested, it disappears from the paper recycling chain.[8] During organic recycling, paper for recycling can be subjected to aerobic (composting), anaerobic or anaerobic–aerobic digestion. Digested and stabilized waste can be used for the reclamation of non-agricultural soils, but first it should be examined by ecotoxicological tests, in order to be sure that it does not contain any toxic substances that can be harmful to the environment and human health.[157,158] Biodegradation of paper products is mostly dependent upon the crystalline/amorphous ratio in cellulose and the presence of lignin.[157,159–161] Amorphous zones of cellulose are more susceptible to biodegradation than the crystalline regions. The relative content of these two components could also vary in the different paper materials, making the process of biodegradation different.[162,163] Different additives in paper are making biodegradation less effective through the inhibition of the enzymatic action.[162] Pinzari *et al.*[164] showed that clay and aluminium can inhibit fungal development during biodegradation of paper more than $CaCO_3$. During anaerobic biodegradation, excessive amounts of calcium can lead to precipitation of carbonate and phosphate, which can affect biomass activity and lead to loss of buffer capacity.[165] Heavy metals originating from printing inks or fillers in paper are not biodegradable and they can accumulate to potentially toxic concentrations. They can disrupt the enzyme activity due to their potential of binding to different groups on protein molecules and replacing naturally occurring metals in enzyme prosthetic groups.[165] When they come from the pulp and paper industry, the most common inhibitors are sulphide, tannins, resin acids, LCFAs and halogenated compounds.[165]

Based on the available literature and results, different biodegradability of paper, depending on the biodegradation conditions, can be seen. According to López Alvarez *et al.*,[166] the presence of paper in municipal solid waste (MSW) has an inhibitory effect and interferes with the efficiency of composting plants, while Zorpas *et al.*[158] concluded that paper can accelerate the maturation of compost. The high C/N of paper may limit its utilization in organic recycling and thus some studies are focused on composting it with different organic waste materials. During the composting process, some undesirable products, such as carbon dioxide, ammonia and VOC compounds, can be found.[157,167,168] The emissions of gases during composting mostly depends upon the composition of substrates and the addition of components responsible for the acceleration of the composting process. Komilis and Ham[168] showed that mixed paper waste (MXP) requires a relatively long time to approximate its full extent of decomposition because of the relatively slow solids hydrolysis. Pommier *et al.*[169] also confirmed that substrate hydrolysis is the limiting step of the paper and cardboard aerobic biodegradation process. Fonoll *et al.*[170] presented the influence of replacing paper for recycling by food waste in the anaerobic digestion process. Results showed that either replacing half of the paper for recycling by food waste or removing half of the paper for recycling had little impact on the reactor methane production. When half of the paper for recycling was removed, methane production was sustained by higher waste biodegradability.[170] The replacement of all the paper for recycling by food waste increased the reactor methane production (37%), while it was estimated that removing all the paper for recycling would reduce the methane production by about 15%.[170] Pretreatment methods can enhance anaerobic digestion of paper for recycling and cardboard, for example, mechanical pretreatments, which are able to decrease the particle size and reduce the crystallinity of the substrate.[23] Sometimes, these methods are high energy-use and do not have a significant influence on anaerobic biodegradation. In order to avoid these problems, biological pretreatment can be used, resulting in low chemical and energy use.[23] Pommier *et al.*[171] showed that shredding strongly affects the macrostructure of the waste, but it does not significantly increase enzyme accessibility to cellulose, as well as the surface bacterial colonization. The positive effect of adding zeolite can be related to the immobilization of microorganisms, which generate a biofilm and thus develop a better environment, increasing the metabolism and improving the yield of the system, influencing the methane production rate.[172] Besides immobilization of microorganisms, natural zeolites have a great capacity for metal adsorption, which is useful for removal of toxic materials that can inhibit the growth of microorganisms responsible for anaerobic processes.[173]

Zheng *et al.*[174] divided paper into two categories according to its lignin content: degradable paper with lignin content of <0.05 g g VS^{-1} (office paper and toilet paper), and refractory paper with lignin content >0.15 g g VS^{-1} (newspaper, coated paper and corrugated cardboard). López Alvarez *et al.*[166] showed that papers presented in Table 8 will never achieve the same level of biodegradation during 45 days, compared to the reference material: microcrystalline cellulose. According to the obtained results, cardboard, kraft paper, newspaper and tissue paper are not suitable for recycling, while white paper (printing) and recycled are compostable.[166]

As already presented and according to the literature available in Table 9, most of the studies are related to organic recycling of paper products, but there is lack of knowledge about the biodegradation of printing inks and their influence on biodegradation efficiency of paper for recycling. In some studies, printing inks are only referred to as toxic components.[166,175] According to López Alvarez *et al.*,[166] cardboard and newspaper are not biodegradable because of the presence of certain chemical components (bleaching agents, printing ink and organic substances used in printing), which may inhibit the biodegradation process, but no results for this were presented. According to EuPIA,[176] the presence of print on a graphic product will not prevent it from complying with the biodegradation requirements and it will not have any major effect on the anaerobic degradability of printed matter in a landfill.

Table 8
Aerobic biodegradation of different paper based products under controlled composting conditions according to López Alvarez *et al.*[166]

Type of paper*	C/N ratio	Biodegradation during 45 days, %	No. days for reaching 70% of biodegradation
Microcrystalline cellulose	-	79.3	-
Printing and writing	177	65.2	65
Cardboard	173	51.7	133
Kraft	5630	36.0	994
Newspaper	422	43.3	309
Recycled	341	61.0	64
Tissue	324	50.1	142

*name given by the author

Table 9
Biodegradation of paper for recycling

Source*	Waste treatment type	Influence of printing inks	Reference
Newspaper	Anaerobic degradation	Yes	[161]
newspaper, cardboard, magazines, plastic paper	Aerobic degradation in compost pile	No	[158]
Hand towels, paper containing mechanical and chemical pulp and high-density paper	Aerobic – controlled composting and full-scale composting in a windrow	No	[157]
Paper bags	Anaerobic degradation – laboratory procedure	No	[184]
Old corrugated cardboard, printed office paper and old newsprint	Aerobic – composting	No	[167]
Old corrugated cardboard, printed office paper and old newsprint	Aerobic – composting	No	[168]
Waste paper	Anaerobic co-digestion		[185]
Copy (CP), news (NP) and box paper (BP)	Anaerobic degradation – batch testing conditions		[186]
Regular paper plates, Earthshell-made plates and commercial biogradable paper	Aerobic – composting in a pile	No	[187]
Waste paper and cardboard	Anaerobic degradation	No	[171]
Filter paper, office paper, newspaper and cardboard	Anaerobic degradation	No	[23]
Paper	Anaerobic degradation	No	[50]
Newsprint, copy paper	Anaerobic degradation	No	[159]
Cardboard	Anaerobic soil environment	UV curable screen printing ink – thermochromic	[177]
Synthetic and recycled paper	Anaerobic soil environment	UV curable screen printing ink – thermochromic	[178]

*name given by the authors

Stinson and Ham[161] studied the decomposition of printed and unprinted newspaper. Based on the amount of methane generated, the results indicated that printing ink did not inhibit the amount nor affected the rate of methane production from cellulose in newspaper.[161] The biodegradation of thermochromic printing inks was conducted by Vukoje *et al.*[177] and Vukoje *et al.*[178] Printed cardboard samples with UV curable ink showed lower values of biodegradation, with a reduction of up to 36%, compared to unprinted cardboard samples after 120 days.[177] For the thermochromic offset print, it was concluded that the biodegradability of the prints mostly depends on the paper substrates used, the penetration of the binder in the paper structure and therefore the thickness of the ink print.[178] Studies of vegetable based polymers and binders, which are commonly used in printing inks, have been carried out by Erhan and Bagby,[179] Erhan *et al.*[180] and Shogren *et al.*[181] Erhan *et al.*[180] studied the biodegradation of ink formulations (soy newspaper ink vehicle, hybrid soy oil newspaper ink vehicle and commercial petroleum newspaper ink vehicle with 18, 25, 27 and 9% by weight of black, yellow, red and blue pigment, respectively). Some differences were found in the abilities of the cultures (*Aspergillus fumigatus*, *Penicillium citrinum* and *Mucor racemos*) to degrade different inks. The ink colour did not appear to affect the degradation rate in the case of the soy oil inks or the hybrid soy ink, but was a factor in the commercial petroleum ink.[180] Shogren *et al.*[181] concluded that highly cross-linked triglycerides with non-degradable linkages (a conjugated system of double bonds) are not biodegradable to a significant extent, while triglycerides (linseed or soybean oil) cross-linked with hydrolysable bonds (carboxylic acid or alcohols), such as esters, remain biodegradable. There are also available data on biodegradation of printed polymer films, in which the reduction of the biodegradation rate was noticed for printed samples.[182,183]

From the presented literature review, it can be seen that the most of the studies are related to different paper types and the influence of different paper components on the efficiency of organic recycling, but almost nothing is known about the impact of printing inks and their behaviour in organic recycling. In addition, it can be concluded that the biodegradability potential of paper for recycling products cannot be predicted, since all the presented results vary from study to study. Despite the limited data, it can be said that the biodegradation efficiency of paper for recycling will be dependent upon the paper substrates used, but the influence of printing inks on biodegradation efficiency will be manifested through their chemical formulation, *i.e.* binder and the presence of toxic components.

Energy recovery

According to the European Commisssion,[40] the energy recovery of paper based products refers to generating energy through direct incineration with the recovery of heat. Thermal processes, such as incineration with energy recovery, pyrolysis, gasification and supercritical water oxidation are the proposed methods.[2] These methods can be used for materials that cannot be recycled and for all kinds of paper products, graphic paper and paper based packaging, regardless of the present inks, coatings, additives and adhesives, which can generally cause problems in material and organic recycling. Most of the studies related to energy production from paper are related to wastes generated from paper production or recycling in paper mills.[3,188–193] According to Vochozka *et al.*,[194] the fillers from paper hamper combustion. Besides fillers, other additives (inks, PVC, PET, Al, polishes and other finalizing products) often raise concerns about the combustion gases and about ash management. Thermal behaviour of paper and cardboard can be studied by thermogravimetric analysis (TGA) and Differential Scanning Calorimetry (DSC).[3,195,196] According to Méndez *et al.*,[188] TGA indicates that the presence of mineral matter and the degradation of cellulose significantly influence their pyrolysis behaviour. In general, weight loss of paper mill waste materials starts at lower temperatures than that of pure cellulose. In the case of wastes from recycled paper, the weight loss continues at temperatures higher than 500 °C due to kaolinite dehydration and carbonates decomposition.[188] Zhou *et al.*[195] showed that CO_2 is mainly produced in the early stage of printing paper and cardboard pyrolysis. According to Zhou *et al.*,[196] printing paper (including blank printing paper and newspaper), with approximately 80% volatile and 10% ash content, can be selected for waste-to-energy thermal conversion. For example, Arenales Rivera *et al.*[3] used thermogravimetric analysis (TGA) for characterization of three different simulating pyrolysis processes, in which the first step (200-360 ºC) corresponds to cellulosic materials, the second step (360-530 ºC) is attributed to plastic materials and the third step (580-720 ºC) belongs to additives of the deinking sludge. Based on the presented results, printing inks may influence the processes of energy recovery through inhibition of combustion, produced gases and ash. However, there are no available data about the influence of different printing inks on the process efficiency and quality of produced gases and ash.

Landfilling

In the past century, landfilling was the main waste disposal method, but it is no longer desirable because of generation of greenhouse gases, leachate, soil contamination *etc*. After disposal of waste in landfills, firstly, aerobic reactions occur within the surface waste, which is in contact with the atmospheric air, but over time, the conditions become anaerobic.[169,197] The aerobic reactions are highly exothermic and they lead to the development of higher temperatures (50-60 °C), which are important for anaerobic biological activity. The initial conditions (temperature, substrate content) for anaerobic activity are influenced by the aerobic reactions kinetics.[169] Taking into account that in landfills, both aerobic and anaerobic reactions occur, we can say that the influence of printing inks on paper decomposition in landfills will be similar to those occurring in organic recycling. The fate of paper in a landfill is a very complex subject, because of a large number of variables (moisture, permeability of cover, temperature, pH and time). It may take a century or more for paper to decompose completely in a landfill. Different authors have studied the decomposition of paper under landfill conditions or laboratory scale conditions, but there is no information about the influence of printing inks on paper decomposition in landfills.[198–201] All the studies showed that papers containing lignin are less degradable than those made from bleached pulp. Also, diapers containing plastic and gels exhibited limited biodegradability.[201] Paper mill industries produce lots of wastes (deinking and paper mill sludge), which is mostly disposed by landfilling or incinerating.[2,3] The problems associated to the landfilling of paper mill and recycling sludge are the large volumes involved and the possibility of hazardous substances leaching into the environment.[2]

LIFE CYCLE ASSESSMENT METHODS

From an environmental point of view, the waste management hierarchy implies that some of the strategies are more appropriate than others, due to their environmental benefits and impacts. Life cycle assessment (LCA) is a methodology that analyses and evaluates the environmental impacts associated with products and processes throughout their whole life cycles: production, use and disposal, by identifying and quantifying energy and materials, as well as the wastes released into the environment.[202] The environmental impact of paper waste management has been studied extensively using the LCA method.[202–204] LCA can be a useful tool for choosing a desirable waste treatment option of paper waste, but in order to get valuable and reliable results, the system boundaries need to be clearly defined, since they can influence the results significantly.[204,205] Villanueva and Wenzel[206] presented in their literature review that the large majority of the analysed LCA scenarios lead to the conclusion that paper recycling is a better option than landfilling or incineration. Despite numerous researches related to the LCA of waste management options for paper waste, there is lack of research about LCA in which the influence of printing inks in paper waste is taken into account. Most of them are related only to pulp and paper waste in general, suggesting different waste management options, such as energy recovery, biogas production, anaerobic digestion or recycling.[175,204,207–211]

Hermann *et al.*[175] emphasized the drawbacks of the obtained LCA results studied for several waste management options for different materials, including paper. The main drawbacks were missing information about the influence of toxic compounds (printing inks) and ammonia emissions during composting, material lamination and thickness, water content and mixing of the compost pile. Dahlbo[211] only indicated that the printing conditions for all the samples included in the LCA study, remained unchanged. An increase in the collection rate also led to an increase in the emissions, except for landfilling and material recycling, which differed from the others by having no energy recovery. Iosip *et al.*[212] showed that higher concentration of contaminants in paper for recycling will result in higher environmental impact due to the increase of energy consumption, the volumes of generated waste and more emissions released into the air and water.

Larsen *et al.*[213] studied the LCA of generic printed matter produced by sheet fed offset printing. The results were used to quantify the possible importance of impact categories related to emissions of chemicals and showed that printing causes significant environmental impacts with a contribution of 41% to the aggregated impact (18% from ink emission at the printing company and 17% from emissions of synthesis chemicals at upstream pigment production). Paper contributes by 31% mainly due to the emissions related to energy consumption. Eco-toxicity and human toxicity were only included to a limited degree or not at all.[213] This study also emphasized that chemical emission-related impacts are generally poorly treated or completely excluded from LCA studies, because of lack of inventory and/or impact assessment data.[213] Larsen *et al.*[213] also indicate that the environmental impacts of chemicals related to printing inks and printing processes may change an energy-related impact profile significantly, at least for product systems with processes with considerable chemical emission (*e.g.* various types of printed matter and textiles).

From the studied literature related to the LCA of paper for recycling waste management options, it cannot be concluded what is the best choice since the results significantly vary with regard to the data collected, the definition of system boundaries, functional units, *etc*. In all the studies, landfilling is the only option, which should be avoided. Despite the lack of information about printing inks in the performed LCA, few studies indicated the importance of taking into account the presence of printing inks in order to get more reliable data about the environmental impact of waste management options, since they can influence the results remarkably. Therefore, good knowledge of the composition and behaviour of printing inks is necessary when considering waste management options.

CONCLUSION

Choosing a particular waste management technology depends upon the quality of the collected paper for recycling. The methods of material recycling proposed for production of recycled paper and cellulose derivatives are mainly aimed at graphic papers, while bioethanol production and organic recycling can be conducted for paper and paper based packaging, which is wet and contaminated with different substances (mainly food), or even for the wastes obtained during production and recycling of paper. Printing inks on paper for recycling may notably influence the quality of the new materials produced, due to the creation of strong chemical bonds to the paper surface during printing, when inks undergo different physical and chemical changes, resulting in different recycling ability.

Ink detachment occurs when chemical and mechanical actions release ink from the fibre. If the prints are not removed from the pulp, recycled paper will not only have poor optical and mechanical properties, but also it can be a problem because of the presence of toxic substances. Thus, it is of great importance to provide a toxicological assessment of the produced materials. Toner (laser, photocopy) and flexographic prints are generally difficult to deink by conventional deinking by the flotation method. Instead, the use of other methods (enzymatic, adsorption and ultrasound deinking) or their combination with the classic method can be conducted. In enzymatic deinking, enzymes can act directly either on the fibres or on the ink film. In addition to the quality of recycled pulp, the effluents from paper recycling processes should also be examined. Studies showed that effluents contain a wide range of toxic components. Enzymatic deinking decreases the pollution of effluents from paper recycling. In addition to lower effluent pollution, enzymes may be a better option as they allow an improvement of optical and mechanical properties of recycled pulp. Up to now, there are no available data about the quality of effluents resulting from ultrasound and absorption deinking. Ultrasound deinking is efficient in deinking toner particles due to an enchainment of ink particles separated from the fibres, resulting in better optical properties. Absorption deinking can be a better option when dealing with digital prints, but special attention should be paid to the choice of appropriate absorbents. In addition, absorption deinking is a promising method when dealing with the removal of mineral oils from pulp suspensions.

Considering that recycling of paper is the most widely studied subject worldwide, the influence of different printing inks on the recycling efficiency is also the most frequently studied and well described in the literature. However, when dealing with other waste management methods considering paper for recycling, there is lack of knowledge about their influence on process efficiency. For the bioethanol production and organic recycling, the only available information refers to the inhibition of enzymatic activity by the toxic substances present in printing inks (fillers, organics and heavy metals). Therefore, printing inks can reduce the efficiency of the processes, but it is not known which types of printing inks affect process efficiency and to what extent. Bioethanol production is mainly studied with regard to the wastes obtained during paper production and recycling, as for organic recycling, most of the available data implies that the biodegradation of paper is dependent upon the presence of crystalline/amorphous celluloses and lignin. The conducted studies report that the type of binder used in printing ink formulations may influence the biodegradation rate. The presence of printing inks may negatively affect the efficiency of producing cellulose derivatives, therefore printing inks must be removed. When the printing ink is removed, the access of the acid to cellulose during hydrolysis is enabled. Most of the studies related to the production of cellulose derivatives involved only mixed office paper printed by non-impact printing (laser, photocopy). For other types of printing inks, there is no available data. Besides the methods mentioned above, thermal conversion with energy recovery, such as pyrolysis and gasification, can be also useful. Most of the studies are related to wastes obtained during paper production and recycling. The presence of printing inks may disrupt the combustion and influence the quality of the produced gases and ash, but the influence of different printing inks on process efficiency is still unknown. Paper for recycling is a valuable material and thus landfilling is the only method to be avoided. The fate of paper in a landfill is a very complex subject because of a large number of variables. It is the only method that is not desirable because of the generation of greenhouse gases, leachate and soil contamination. LCA can be a useful tool for the selection of a desirable waste management method, but only if the system boundaries are well defined. The main drawback of the LCA studies conducted up to now is the lack of information related to printing inks used for calculations of environmental impacts.

Comparing all the possible waste management methods targeting paper for recycling, the most significant influence of printing inks on process efficiency is known to be in the case of classic recycling for the production of recycled paper, but when dealing with other methods, there is lack of available data. In these methods (bioethanol production, organic recycling and energy recovery), the efficiency of the processes is mostly influenced by the types of paper substrates and their components (*i.e.* presence of lignin, fillers *etc.*). Based on the literature presented, in all of these waste management methods, the printing inks play an important role, thus when researchers are dealing with some new recovery method or new materials, they should always take into account the presence of printing inks.

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