https://doi.org/10.2352/ISSN.2169-4451.2017.32.435

Printed Electronics and 3D Printing as New Manufacturing Technologies - New Opportunities For Bio-based Materials

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Abstract

Printing has made significant impact on our society and daily life, particularly through process improvement (high speed, high volume) and technology advancement (e.g. digital printing, personalized printing). However, in recent years, the spreading of internet has created significant impact on printing, leading to the decline in demand for graphic printing. Despite these challenges, printing as a well-established technology is now being explored for new applications including printing electronics and 3D printing. With these manufacturing technologies, new opportunities/applications for bio-based materials, including lignocellulosic materials are being explored, as reviewed in this report.

Introduction

Printed electronics (PE) is being increasingly explored for many applications such as OPV, OLED lighting/display, energy storage devices, RFID, etc., and a few of them have even been commercialized. However, the commercial utilization of the PE technologies is mainly on conventional electronic substrates (silicon/glass wafers) and a small portion is on high-quality coated plastics/paper. With an increasing demand for sustainability, PE is attracting interests in packaging (use paper/board-based materials) and also as a tool to market/sell more products by being interactive (touch sensing) and to monitor the movement of packaging and the quality of packaged goods (RFID type of biosensors). Although paper-based packaging is well positioned to meet the requirement of sustainability, applying PE technologies directly onto paper substrates to render the smart functionalities for the packages faces some technical challenges.

3D printing, as an advanced printing technology, has been used to produce prototypes and molds in industry for many years and is now gaining increasing interest, due to its expansion into consumer products markets as well as the exploration of potential applications in manufacturing large architectural structures. Forest industry products based on lignocellulosic materials are already being prototyped using 3D printing. There is also an increasing interest using wood-based materials as feedstocks for 3D printing to impart unique appearance for printed products and meet the demand for sustainability from the consumers. This could become part of the transformation of the forest industry from the production of commodities to the production of high value-added specialty products.

In the PE part of this work, we will discuss the potential opportunities for PE on paper-based substrates for creating sustainable and functional packaging as identified through market analysis and value proposition assessment. Our strategies on how to overcome some technical challenges for printing electronics on paper substrates such as high roughness, non-uniformity of surfaces, low vapor and oxygen barrier will be presented. One of the promising concepts based on paper and board that are being explored at FPInnovations will be described. These examples will be used to show how to create sustainable and functional paper-based packaging with PE technologies.

In the 3D printing part of this work, we will summarize the worldwide effort on exploring potential applications of bio-based materials: compounding wood-containing bioplastics for making interesting objects, bio-printing with cellulose nanomaterials for biomedical applications, and developing wood-containing feedstocks and new production concept for large-scale additive manufacture of advanced wood-based 3D structures. We will discuss the technical challenges existing in developing feedstock formulations that will match bio-based materials with the most appropriate 3D printing technology. Through market analysis and technology assessment, FPInnovations has created a vision on how the forest industry can utilize the 3D printing technologies for creating new applications and product opportunities.

Printed electronics: opportunities for cellulose products

Paper vs. plastic as substrates

Plastic films are generally considered to be superior to paper for printed and flexible electronics, due to greater smoothness and very low porosity. However, plastic films have drawbacks, notably high stretch and sensitivity to heat. On the other hand, cellulosic substrates are seen as too rough and too sensitive to moisture.

Appropriate coatings can bring paper surfaces to the same smoothness as all but the most specialized plastic films. The second perceived drawback for cellulosic substrates is the moisture sensitivity. Both experimental work and commercial products involving barrier coatings show that this problem is also resolvable.

One critical need is virtually flawless printing. A break in a printed line that has no visual impact on a high quality print could destroy the operation of a printed electronic circuit. As one author put it, “You can cheat the human eye but not the conductivity” [1]. It is often stated that plastics are superior to paper in this regard, but this may not always be true.

Paper, board, and label stock already act as simple carriers for pre-assembled electronics, such as silicon-based RFID chips. If paper is to act as the actual substrate for printed electronics, can off-the-shelf paper products be used, or is there a need to develop new paper grades? Another development is the assembled product, where the paper is the base layer in a manufactured sandwich structure.

PE applications on paper and cellulose-based substrates

There is potential for three different types of forest products in the field of printed electronics. The first comprises existing paper products, with little or no modification. Some off-the-shelf coated papers or label stocks have the appropriate smoothness and porosity to accept conductive inks, or even to allow multi-layer printing of circuits. This could also include film products such as cellophane.

The second type consists of paper surfaces specifically developed for printed electronics. The third type consists of specially developed cellulosic derivatives, of which various forms of nanocellulose are leading possibilities, as discussed below. Trnovec et al. [2] of Chemnitz University of Technology, Germany, compared paper as a substrate to a "standard" Mylar surface. They noted the advantages of paper including price, its much wider availability for a variety of end-uses, and its greater environmental friendliness. They showed that papers with excessive penetration of the conductive inks were not suitable for printed electronics, and suggested that a pigment-free, low porosity barrier layer coating may be preferable for printed electronics. The Swansea University group [3] used flexography to print lines of a conductive carbon ink on regenerated cellulose film, and determined that the properties of this substrate were adequate. The authors noted the advantage of both a biodegradable substrate and ink.

A specialty paper manufacturer, Felix Schoeller of Germany, has introduced three new products for printed electronics. These have their roots in traditional photographic paper. All three products start with conventional paper basesheets. The manufacturer applies a barrier layer to both sides of the sheet (e.g., a polymer resin), followed by a functional surface layer. A Cambridge-based manufacturer, PEL Ltd., offers an "electronic grade paper", with a nanoporous surface layer. This is claimed to withstand sintering temperatures of at least 150° C, and possibly higher for very brief periods.

Nanocellulosic substrates

Most plastic films have the smoothness and low porosity required to print very finely detailed electronic circuit components. However, these films have high coefficients of thermal expansion. Recent work [4] has shown that conventional polymer films either laminated or filled with nanofibrillated cellulose (NFC) have an extremely low coefficient of thermal expansion. Films made from cellulose nanocrystalline (CNC) also show very low coefficients of thermal expansion [5]. Composite sheets made from NFC and up to 80% mineral fillers gave very smooth surfaces, with properties (including dimensional changes with humidity) similar to those of a reference Mylar sheet [6].

Functional, structured, and assembled/printed devices

In 2011, Acreo of Sweden commercialised what they term an "electronic barcode" – a fully printed, RFID technology that does not require a silicon chip [7]. Few details were given, other than that the technology can be used on virtually any substrate, and can be overprinted with conventional optical barcodes. It may be assumed that these tags carry relatively little information. For many years, the Nordic pulp and paper giant UPM-Kymmene had a commercial RFID division that supplied RFID tags that were applied to ordinary label stock. This division of UPM was acquired by Smartrac in 2011, a Netherlands-based supplier of RFID tags and transponders [8]. The stated goal of Smartrac in this transaction was to add UPM's packaging RFID expertise to their portfolio.

Another specialized application is the flexo printing of wallpaper with conductive silver ink by the Centre Technique du Papier (CTP) in France [9]. The goal of the project was to permit the construction of rooms that would block radiofrequency waves, to create "wi-fi" free zones. Presumably, it would also be of interest to use conductive inks other than those based on silver. A group at MIT has successfully developed solar cells on plain paper and fabric. In 2012, MIT was awarded $10 million by the U.S. National Science Foundation [10] for a five year project, under the title "An Expedition in Computing for Compiling Printable Programmable Machines". Proof-of-concept work has already produced miniature working robots based on single sheets of paper or Mylar film. The Chemnitz University group [11] produced photovoltaic cells on a paper substrate. A glue layer was printed onto a coated paper, followed by transfer of a conductive zinc layer. The photoactive layer was printed by gravure, followed by flexo printing of PEDOT:PSS.

"Smart" sensors, bioactive paper, and printed medical diagnostic paper

Printed, functional papers may also be considered as part of the same general field of development as printed electronics. This may include electronic circuits that act as sensors, or printed surfaces that act as chemical/biological sensors or cleansing agents. Several have already been described in this review, including strain gauges**,** conductivity-based H_2S sensors, and conductivity-based moisture sensors. Canada's Sentinel Bioactive Paper Network [12] was active from 2005 to 2015. Output from this network has included the application of printing technology to the preparation of bioactive devices.

Conventional transistors can be used as chemical sensors, warning of problems such as (for example) excessive exposure to heat for food or pharmaceuticals, or warning of the presence of hazardous materials. VTT, Finland, described two printed sensors for detecting tainted food [13]. Since hydrogen sulfide is a by-product of deteriorating meat, both devices are based on the reaction of H_2S with silver. A simple version uses the colour change of a printed silver line to black silver sulfide. A more complex version is based on the conductivity drop between silver and its sulfide. VTT is also developing other paper-based diagnostic materials. Some may contain simple, single-phase diagnostics (e.g., reaction with a single chemical reagent), while more complex devices involve microfluidic separation. Kahn [14] showed sensors printed on plastic film and coated paper that were sensitive to materials such as water, ammonia, and hydrochloric acid. Jabrane *et al.* [15] (part of Canada's Sentinel Bioactive Paper Network), used a small laboratory flexo press to produce bioactive paper capable of destroying E. coli bacteria. A key conclusion was that the bacteria-killing virus survived the printing process.

A market study was conducted by FPInnovations in 2012 to identify market niches where paper and paperboard have an advantage over plastic substrates, and the Canadian forest sector may have an advantage or opportunities over their competitors. The study showed that, despite past significant international R&D investments, challenges remained with respect to design, production and functional materials and that there were few commercial printed electronics available in the marketplace.

Fig. 1 100% paper-based and using standard printing to add a conductive layer to improve performance & lower cost

However, the research also showed that the world market for printed and flexible electronics was projected to increase from $1 billion in 2012 to $25 billion by 2022.

After completing the detailed study of market potential and technologies FPInnovations decided to focus on 'smart' packaging applications (e.g., the use of PE to provide brand enhancement, interactivity and traceability to packaging products) and active packaging. The concept and the technical progress for printed susceptor will be discussed in detail here.

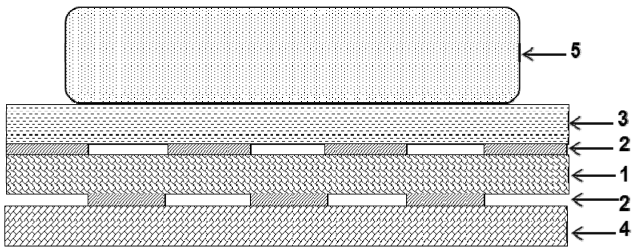
Printing a duplex microwave interactive susceptor structure on cellulose-based substrates

Active microwave packaging is a class of microwaveable food containers, including bags and boxes that use microwave interactive materials (susceptors) to enhance cooking performance. The susceptor layers are the key to active microwave packages because they convert microwave radiation to heat. These are made of metallized plastic films laminated or inserted inside the packages. Although such metallized plastic film susceptors are widely used in the microwave food packaging market, this technology still has performance and recyclability/compostability issues. For example, the metal layer is typically 5 to 10 nm thick, which cannot produce enough heat for some applications. However, if the thickness increases, this metal layer reflects the microwave radiation rather than converting the microwave energy to heat. Another technical issue for the metal film is that it easily breaks down when it is heated, due to its ultra-small thickness. Although the breakdown of metal films can be designed as a safety feature to avoid overheating, it does limit the upper temperature that the metalized film can reach.

Novel susceptor fabrication technology derived from conventional material deposition techniques such as printing and coating onto cellulosic substrates would help this market grow, as it would provide a potentially low-cost and green alternative to metallized plastic films. However, although the concept of printed susceptors has been patented, it has not yet been commercialized. The reason is that printed susceptors still have performance issue, particularly the potential safety issues caused by "runaway heating", which can result in the package catching fire. One possible solution to this critical issue is to print smaller susceptor areas interspersed with areas that do not interact with the microwave radiation. The size of the microwave non-interactive pattern is recommended to be the same size as the interactive pattern, to avoid the occurrence of arcing on two adjacent interactive patterns. However, this reduces the total microwave interactive surface area, and thus reduces the heating capability of the susceptor. Overall, one key criterion for good susceptor performance is the ability to reach a safe upper temperature limit at which a steady state absorption of microwave energy occurs. Recyclability or compostability of susceptors could also be an important asset.

Our solution is to produce a 100% paper-based susceptor using a standard printing press (Fig. **1**). The design of the duplex susceptor package is shown in Fig. **2**. The middle ply as a support layer for the microwave interactive structure may be a low basis weight (less than 100 gsm) paper or a high basis weight (larger than 100 gsm) paper. The microwave interactive material is patterned on both sides of the substrate and spatially staggered. We call this a duplex susceptor. The microwave interactive material is isolated into defined patterns but still covers the whole surface of the cellulosic substrate. The duplex susceptor may comprise an electroconductive or semiconductive material, for example, a metallic ink/paste, an organic ink paste, or any combination thereof. The top ply food barrier layer may be a cellulosic film with high grease resistance or a printed layer of any food safety grade material for high temperature (around 100°C to 300°C) such as polyester, silicone, etc. The bottom ply as the outer layer for the package may be paper, paperboard or another acceptable non-microwave interactive cellulosic material.

The pattern for the duplex susceptor mentioned above may be in the shapes of squares, circles, loops, hexagons, rectangles, octagons, and so forth. The sizes and gaps of these patterns may be in the scale of microns to centimeters, only if the shape and positioning provide sufficient radiation to reach the desired temperature.



1. a middle ply; 2. susceptor layer; 3. a top ply; 4. a bottom ply; 5. a food item

Fig. 2 A cross section of the design of the duplex microwave susceptor package

The uniqueness of this work is that the duplex design maximizes the surface coverage of the microwave interactive material, and the material is still confined in discrete patterns. The larger surface coverage means more energy converted and the energy is evenly distributed over the surface compared to the single side, discrete pattern susceptor. The discontinuities in the microwave interactive layer could confine the random flow of eddy currents induced from the interactive particles, reducing the risk of forming hot spots thus preventing package ignition. Fig. **3** shows that the duplex susceptor heats up a tube of silicone oil to a higher temperature at a faster speed compared with the single side, solid susceptor and the single side discrete pattern susceptor.

The patent-pending printed paper susceptor technology[16] from FPInnovations has been tested on a commercial press (Fig. **4**). By implementing this technique, customers of paper producers, the converters, can have increased control of pricing of final products by simplifying supply chain. FPInnovations is looking for partner(s) to further demonstrate this technology at converting plants.

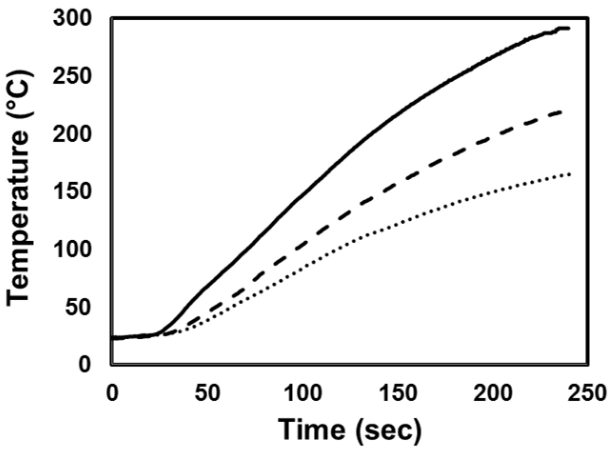


Fig. 3 Time vs. Temperature recorded from a tube of silicone oil heated by different susceptors. Single sided, discrete pattern susceptor (Dotted line), Single sided, solid susceptor (Dashed line), Duplex susceptor (Solid line).

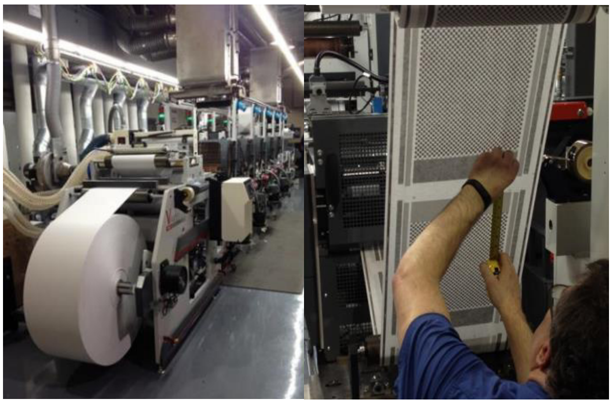


Fig. 4 Tunable heating levels of susceptors by adjusting printing conditions on site

3D printing and bioprinting: Opportunities for lignocellulosic materials

Considerable research is ongoing to use wood and forest products as 3D printing feedstocks. These efforts span the range from small scale, highly specialized value-added bioprinting to large scale building manufacturing.

Use of lignocellulosic materials in powder and FDM printing

3D printing of thermoplastic filaments has grown considerably in recent years, and there is already a demand for "green" substitutes for petroleum-based thermoplastics. Polylactic acid (PLA) is currently the principal biomaterial for 3D fused deposition modeling (FDM) printing, but has issues of low strength, low flexibility, poor thermal stability, and poor printability. Bio-based materials extracted from forest products can be tuned to address these issues. However, their market potential and value need to be established. Significant efforts worldwide in recent years have led to the production of large quantities of bio-based materials in pilot and demonstration plants. However, technical challenges exist in developing feedstock formulations that will match wood fibres, cellulosic materials or lignin with the most appropriate 3D printing technology.

Work at FPInnovations and elsewhere has shown that these problems can be solved, at least at the scale of proof-of-principle. FPInnovations and Emily Carr University of Art and Design (Vancouver) collaborated on the exploration of 3D printing of powdered lignin using powder-binding technology. The objects in Fig. **5(a)** are composed of lignin plus binder. This work is being continued by FPInnovations, and includes the development of wood-based filaments for 3D FDM technology. The objects shown in Fig. **5(b)** were made from filaments composed of thermoplastic polymers containing lignin filler and fibre materials.

Wood containing thermoplastic filaments, supplied by non-systems manufacturers, for fused deposition printing are already on the market. In 2012, CC-Products created Laywoo-D3 [17], containing up to 40% recycled wood fibre, combined with a thermoplastic polymer binder. ColorFabb, produced by Helian Polymers of the Netherlands, offers WoodfillTM, containing 25-30% milled wood fibre in a thermoplastic resin [18].

Use of lignocellulosic materials in bioproducts

As already noted, 3D printing is being actively developed by manufacturers of medical devices. In one recent life-saving case, a bronchial "splint" was 3D printed to help expand the bronchial tubes of an infant. The device was made out of a biopolymer (polycaprolactone), and is expected to be gradually absorbed into the child's body, allowing its own pulmonary system to take over [19]. The ease with which cellulosic derivatives form stable dispersions and gels in aqueous media provides an excellent opportunity for new bioproducts. While obviously removing large amounts of water presents a drawback, nevertheless, this is an active area of world-wide research.

Using a bioprinter, a group in Wales prepared 3D-printed wound dressings, using nanofibrillated cellulose (NFC) supplied by the Norwegian Research Institute PFI [20]. They described their resulting material as strong, which could be

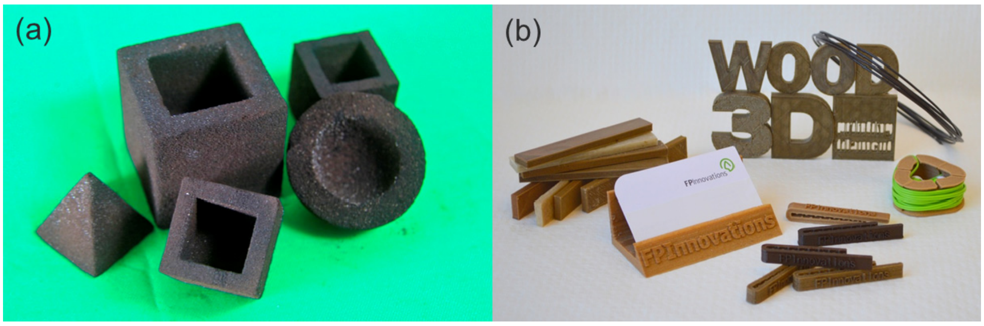


Fig. 5 Developing lignocellulosic-based feedstock for 3D printing at FPInnovations (a) Prototypes made using the powder-binding technology; (b) Prototypes made using FDM technology

kept under moist conditions and possessed inherent anti-microbial activity –perhaps the result of surface modification during the oxidative process used to prepare the nanofibrils. The Swansea group also used more conventional materials to build a 3D scaffolding that effectively formed a collagen-like structure [21].

A different approach was taken by a group in Sweden, involving the use of a dissolving pulp (in an ionic solvent) as the cellulose source [22]. The printed cellulosic gel was then immediately coagulated by overprinting a layer of water. Successive layers can then be built up. Although the solvent that was used in that particular work is rather exotic, there exists a very extensive chemistry for dissolving and then regenerating cellulose which could be applied.

3D printing large scale wood products

3D printing of building components and even entire buildings has gained considerable attention around the world. So far, workers in the field tend to use concrete or related materials in extrusion type printers. At the same time, forest industry researchers are examining the potential for using wood products in large scale 3D printing construction.

Researchers at the University of Sydney (Australia) recently received funding to develop "micro timber" [23]. The process, not fully described, claims to use wood products as one element in 3D panel printing. Potential ingredients include hardwood and softwood fibres, wood flour, and even wood waste. Other details (including binders) have not yet been disclosed.

We can compare this work to concrete extrusion printing, in which the concrete could either contain wood fibres as a reinforcing component, or where the main solid portion of the structure would be wood-based. Concrete as an aqueous slurry is also subject to the requirement that the structure rapidly become self-supporting; certainly before the next layer is printed.

The Oxman group of the MIT Media Lab is dedicated to both the art and science of 3D printing. In particular, they have a special interest in both permanent and biodegradable structures. A very recent publication on "water-based engineering" [24] described printing composites made from chitosan and cellulose, among many other materials.

Another variation on this is the use of wood products (e.g., fibres, wood flour, or even wood chips) in a form of powder-bed printing. Just as in powder bed printing with sand or other particular material, the wood product would form the "powder bed", and complex structures would be built up by printing a suitable adhesive, layer by layer. Researchers at the Technische Universität München demonstrated a truncated cone generated by 3D printing with wooden chips as bulk material and gypsum, methyl cellulose, sodium silicate and cement as binder [25].

Through market analysis and technology assessment, FPInnovations created a vision on how the forest industry can utilize the 3D printing technologies (Fig. **6**). Further technical progresses will be reported in the future publications.

Summary

Creating new sustainable products using PE technologies and developing "green" feedstocks for 3D printing are in the early stages at FPInnovations and within industry. These new products currently represent niche market opportunities. These projects have resulted in a number of technology platforms, products and processes that represent ready (or near-ready) opportunities for the printing, packaging, medical sectors, etc. More work in the lab, pilot-scale trials with engagement of end-users are required to improve the technical readiness level of these new technologies/applications.

Fig. 6 FPInnovations' vision on 3D printing for forest industry

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