Functional Inks and Indicators for Smart Tag Based Intelligent Packaging Applications

Maria Smolander  
Kaisa Vehmas

Liisa Hakola*
VTT Technical Research Centre of Finland Ltd

ABSTRACT

Smart Tags are functional, information transmitting elements that combine 2D barcodes and environmental sensing into a cost effective tag that can be attached to e.g. product packages, where additional elements should not increase the product costs significantly. Important feature of these Smart Tags is that they can be attached to products in very high-speed production lines, which makes them suitable to fast moving consumer goods. Because the Smart Tags are sensitive to environmental conditions, they are dynamic, but they also enable context aware services as each of them can be unique. The enabling technologies behind these Smart Tags are i) 2D barcodes and ii) functional inks, such as thermochromic and photochromic inks, and iii) printed visual indicators. In this paper, different ink and indicator technologies are used to build Smart Tags. Both commercial inks and developmental grades are in focus. Furthermore, it is evaluated if these tags can be detected by mobile phone reader.

KEY WORDS

smart, tag, intelligent, packaging, barcode, indicator

*Liisa Hakola  
Corresponding Author  
liisa.hakola@vtt.fi
1 INTRODUCTION

Packages usually contain only a limited amount of information about the product it contains. In the food value chain, there are several points, where the product related information would be valuable to be communicated to both directions. By utilizing tagging technologies, such as Smart Tags, it is possible to provide applications that include recording, storing and transmitting unique dynamic and context-aware information about the food products in addition to other statistic information about the food product, e.g. origin of ingredients, production conditions, producers, stakeholders, authenticity, and transportation conditions. The information received from this type of intelligent packaging can be communicated to users through digital services accessed by mobile phone. The information could even trigger the function of active packaging i.e. means to control the quality of the packed product by using electronic functionalities connecting the monitoring and active packaging.

1.1 Smart Tag technology

The concept of using functional inks in enhancing dynamic nature of 2D barcodes is Smart Tags. Smart Tags has been first introduced by TagItSmart project funded by EU’s Horizon 2020 instrument [1, 2]. Smart Tags mean visible or electronic markers, such as 2D barcodes and NFC tags (Near Field Communication), with environmental sensing functions (functional ink, sensors, indicators) combined with software intelligence (machine vision, user information, location, etc.). Previous features provide context aware services to the end users and enable the connectivity to the Internet of Things (IoT), in a way that hasn’t been possible for legacy products before.

From a technology perspective Smart Tag consists of i) a marker that provides machine reading possibility, (unique) identification of an item and linkage to online services, ii) an environmental sensing area that changes the code reading results dynamically when surrounding environmental conditions change, and iii) associated digital service, such as webpage or cloud repository. This paper focuses on a Smart Tag version that consists of a 2D barcode (marker) combined with sensing function resulting from use of functional inks or indicators. When some parts or areas of a 2D barcode are printed with the functional ink, the scanning result of the code changes dynamically when the state of the functional part changes. Combination of 2D bar code and functional part can be achieved

![Figure 1: Some options for using functional inks as part of QR codes. a) (red and grey) cells printed with functional inks, b) (green) area printed at the bottom of the code with functional ink, and c) OCR (Optical Character Recognition) (yellow letter “C”) combined with QR codes.](image-url)
in different ways as pictured in Figure 1. When conditions of the surrounding environment change, such as ambient temperature, lighting or humidity, parts of the barcode appear, disappear or change color. This causes the visual appearance of the code change and the scanning result to change accordingly. Besides encoding the 2D bar code, the scanning software can also interpret different colors i.e. color of the functional area. Thereby, the environmental conditions have an effect on the scanning process and resulting digital service. The services can also take into account the other user and context related data, such as user profile and GPS location of the smartphone. These context-aware features will further improve the value and quality of the services provided.

TagItSmart research project has piloted several use case scenarios for Smart Tags in real-life environments [1, 17, 22, 23]. In one use case, the Smart Tag gives information on life-cycle of the item and indicates a correct consumption temperature as well as gives advice on recycling of the package. The consumer can also be rewarded after recycling is completed. In another scenario, the product authenticity is verified with the Smart Tag based on an ink reacting to mobile phone flashlight. In a third scenario, the price of the product is determined at the cash register based on state and traceability history of the Smart Tag. The piloting activities during TagItSmart project also spanned outside food packaging sector with cases dealing with medical distribution in hospitals, with manufacturing domain, and with home appliances use condition monitoring.

Consumers, the potential users of these services, evaluated the TagItSmart use cases to be very interesting for them and emphasized their interest in an interactive relationship with service the provider [7]. In addition, consumers are willing to use these kinds of novel services if they feel that they get additional value from them. Retail store attitudes on intelligent package solutions have been investigated in one recent study [24]. Reduction of product loss to spoilage was considered a significant benefit when quality indicators act as dynamic use-by-dates. However, use a dynamic use-by-date solution could lead to too easy discard of an item by consumers. If one package shows spoilage, consumers might consider that all packages of this specific product are not in proper condition.

1.2 2D barcodes

One technology building block of Smart Tags are 2D barcodes that have been available for almost 30 years, but during the recent years, their popularity has increased due to the growth in smartphone ownership with capabilities for 2D barcode detection [3]. 2D barcodes are at the moment used for traceability, information, entertainment and even for payment solutions. 2D barcodes, such as QR Codes and Data Matrix codes consist of black and white squares, called cells. They provide a large information capacity, even up to 7336 numbers or 4464 alphanumeric characters per code. A sophisticated error correction algorithm is included, which means that information is readable even if up to 30 % of the code is destroyed. 2D bar codes can serve as a link to a database similar to linear bar codes, but they can also serve as an independent database. The physical size of the code is scalable without affecting the information capacity meaning that the cell size of a particular code can be scaled. QR Codes can be easily detected and decoded with many devices, like mobile phones [4], thus providing a link to a digital media and additional information. 2D barcodes can be printed on many types of substrates with regular printing inks and processes. Printing even on edible products is possible with suitable inks [5, 6]. 2D bar codes can have many uses in intelligent packaging.

Chen et al. have reported a mobile solution based on 2D barcode technology for traceability of pork [7]. This solution is based on static QR codes without additional functional elements. For traceability purposes also RFID technology has been used,
but also without environmental or headspace sensing functions [8]. There are some solutions where linear barcodes are used together with sensing functions. In these cases the appearing color makes the barcode unreadable, but does not give any additional information to users [9].

Yuan et al. have reported an integrated QR code and biosensor for food supply chain monitoring based on silver-enhancement self-assembly [10]. In this solution, areas of the QR code are missing until target detection. Since the developed code is based on antibodies and gold nanorods, it is not as cost-effective solution or suitable for high-speed production as the Smart Tag concept based on functional inks. However, due to use of antibodies more advanced detection than environmental parameters is possible, such as bacteria.

Wang et al. have used infrared watermarking together with QR codes for anti-counterfeiting features [11]. The implicit graphic included during code creation can be detected under infrared light. The proposed method is compatible with the existing workflow of printing without using special inks. The black ink is used for imaging under IR light, and the code is constructed by using a sophisticated algorithm.

Outside intelligent packaging, Burklund et al. have reported a colorimetric assay combined with QR code for screening volatile biomarkers. In the solution, the reagent sensitive to e.coli bacteria is placed on defined regions of the QR code. In the presence of the analyte, the reagent becomes visible and affects the scanning result by mobile phone. Although the application area is outside intelligent packaging and colorimetric assay based on microfluidics more complicated than functional inks, the principle idea of combining QR code with color changing areas is similar to Smart Tags [12].

1.3 Functional inks and indicators

The second technology building block for Smart Tags are inks or indicators that provide environmental sensing function. The use of functional inks reacting to different environmental conditions enables creation of novel digital services for the consumers and other stakeholders [8]. Functional ink and indicator technologies make it possible to create dynamic 2D barcodes i.e. Smart Tags that enable changing the achieved information and its meta-information content due to the changing environmental conditions when scanning the codes by mobile phone [5].

The most popular and readily commercially available functional ink technologies are thermochromic and photochromic inks. Reversible thermochromic inks change from one color when in their cool state to translucent when in their warm state. Irreversible thermochromic inks change permanently from transparent to a colored state when heated. Photochromic materials change their color when the intensity of incoming light changes. Other commercially available ink technologies include invisible fluorescent inks that can be seen under UV or IR light, phosphorescent inks that glow in the dark after exposure to a source of light, hydrochromic inks that change color after contact with water, and touch’n smell inks that release aroma when rubbed with a finger, among others.

Most of the commercially available functional inks are available for analog printing methods, such as flexography and screen printing. There are mostly fluorescent inks available for digital printing methods, such as inkjet printing. The advantage of using digital printing methods is a possibility to use unique codes with tailored content.

Indicators with application to food packaging are time temperature indicators, oxygen and integrity indicators, and freshness indicators. Indicators can be based on mechanical, chemical, electrochemical, enzymatic or microbiological changes as recently reviewed e.g. by Ghaani et al. VTT (Technical Research Centre of Finland) has developed several printed indicators for monitoring e.g. food quality indicating volatile compounds, humidity and oxygen, which may have a crucial effect on the product quality (Figure 2). Most of them are compatible with inkjet printing.
technologies [5, 14, 15]. These are ink-based technologies similar to functional inks, thus enabling printing these chemically reactive indicator inks together with QR codes to create Smart Tags for supply chain management, product authentication or quality indication.

1.4 Smart Tags for intelligent packaging

The most popular intelligent packaging technologies are sensors, indicators, barcodes and RFID (Radio Frequency Identification) that can be implemented with available technologies. Smart Tag technologies combine at least two of these technologies. Sensors are devices that detect and respond to selected inputs from the physical environment, and the output is generally a signal that is converted to human-readable display [17]. Indicators are sensors that give some information about the occurrence of a certain condition (e.g., presence of a chemical substance or reaching a temperature-threshold) based on optical reading, such as color change [17, 20]. The type of Smart Tags described in this paper open up opportunities for more advanced context-aware IoT services on packages thanks to their affordability [17]. There are, however, still development needs with the technology. In many cases, it would be necessary to control the speed of color change reaction. This would be achievable by modifying the printing layout and/or the ink chemistry, and by using e.g., protective coatings or laminations to decrease the reaction speed. Another challenge is that some of the ink chemistries require printing of multiple layers for sufficient darkness that can be scanned by mobile phone. Also, this could be tackled by modifying the ink chemistry, e.g., by increasing colorant concentration.

The goal of intelligent packaging technologies is to provide means for controlling packed product quality, to provide more convenience to consumers, to market and brand the products, and to control counterfeiting and theft [18]. Poyatos-Racionero et al. have introduced the possibility to use intelligent packaging even to reduce food waste with freshness indication [21]. Smart Tags are another suitable solution for intelligent packaging at high-speed production line. The technology has been evaluated with the food packages that emphasizes the ability to sense or measure and further communicate an attribute of the packaged food product, the atmosphere inside the package, or the environment [18, 19]. The benefits of using Smart Tags include consumer engagement through personalized and

Figure 2: Printed indicators for oxygen (a) and for ethanol (b).
context-aware services, direct personal contact for brand owners with consumers, increase in product quality and safety, product authenticity and integrity, as well as reduction of waste [2]. This will also lead to increasing transparency in the food value chain and consumer trust towards different food products. Besides low-cost manufacturing process, one technological advantage of Smart Tags is the use of mobile based code scanning. Thereby, the user does not have to understand what the different colors on Smart Tags mean. The mobile app interprets the color in a correct way and returns the service, accordingly.

1.5 Smart Tag objectives in the scope of this paper

The objective of this paper is to use different ink and indicator technologies to print Smart Tags and to evaluate if these tags can be detected by mobile phone reader in order to build context-aware IoT services for end users along the packaging value chain, all the way from brand-owner to consumer. Since functional inks and indicators are one of the enablers for Smart Tag creation, they should be evaluated for their capability to produce 2D barcodes that are the second enabler for Smart Tags.

Since Smart Tags are targeted to be used by consumers, retail stores and other end users with mobile phones, it is important to understand if functional inks and indicators can provide areas that are dark enough to be detected by mobile phone reading. A representative set of different ink and indicator technologies were chosen for the experimental part in order to evaluate different chemistries, both commercial and developmental grades. Since most of the commercial inks are for analog printing methods, the inkjet printable inks were formulated from pigment or dye dispersions. The indicator technologies used were developmental grades, since no commercial grades are available as liquid samples - only as ready-made indicator tags.

Since Smart Tags are printed features, the choice of which printing technology to use plays a significant role. Flexography and screen printing are analog printing methods where a physical master is used for generating prints on substrate. Analog printing methods provide high throughput and compatibility with multiple ink and substrate materials, but are not suitable for printing smaller series economically, such as unique tags. In digital printing the print is generated from a digital file, thus enabling making even individualized prints economically. The drawback specifically with inkjet printing is lower speed than in analog printing, and high dependency on ink-substrate compatibility, thus limiting the availability of suitable materials. However, both analog and digital printing technologies can be considered suitable methods for printing Smart Tags. The choice of the printing method to use has to be made based on application and end use requirements. In the experimental part both analog and digital printing will be addressed.

Different packed product categories have different requirements for Smart Tags and what environmental conditions should be monitored. For example, in some cases information if a product has been stored at incorrect conditions, or if the package is still intact, are required. This means that irreversible inks that permanently change due to improper handling conditions are useful. In other cases, when the purpose is to show e.g. correct usage conditions or provide authenticity information, reversible inks are a more suitable solution. Thereby, both reversible and irreversible inks were selected for the experimental part.

2 MATERIALS AND METHODS

Eight different functional ink and indicator technologies were evaluated. These included both commercial inks for flexography and screen printing, as well as developmental grades for inkjet printing. In addition, the indicators evaluated were developmental grades. The inks and indicators were:
Thermochromic inks:

1. Reversible thermochromic flexographic ink: WB FLEXO GREEN TO CLR FC 08C from Chromatic Technologies Inc. (commercial ink) (TC1)

2. Reversible thermochromic inkjet ink: formulation based on Chameleon pigment dispersion from LCR Hallcrest, temperature 47 °C (developmental grade) (TC2)

3. Reversible Chameleon WB flexo ink slurry, color red, temperature 47 °C from LCR Hallcrest (commercial ink) (TC3)

4. Reversible Chameleon WB flexo ink slurry, color black, temperature 31 °C from LCR Hallcrest (commercial ink) (TC4)

Photochromic inks:

5. Reversible photochromic flexographic ink: UV flexo red photo from Chromatic Technologies Inc4. (commercial ink) (PC1)

6. Reversible photochromic screen ink: Chameleon water based screen ink RED from LCR Hallcrest (commercial ink) (PC2)

7. Reversible photochromic inkjet ink: formulation based on photochromic RED#19 pigment from LCR Hallcrest (developmental grade) (PC3)

8. Irreversible photochromic inkjet ink: formulation based on dye based Lumi Inkodye RED (developmental grade) (PC4)

Indicators:

9. Irreversible oxygen indicator flexographic ink: proprietary formulation based on a reduced redox dye (IN1)

10. Irreversible humidity indicator inkjet ink: proprietary formulation (IN2)

<table>
<thead>
<tr>
<th>Ink No.</th>
<th>Code</th>
<th>Type</th>
<th>Reversible/irreversible</th>
<th>Commercial/developmental</th>
<th>Printing method</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>TCI</td>
<td>Thermochromic ink</td>
<td>Reversible</td>
<td>Commercial</td>
<td>Flexography</td>
</tr>
<tr>
<td>2</td>
<td>TC2</td>
<td>Thermochromic ink</td>
<td>Reversible</td>
<td>Developmental</td>
<td>Inkjet</td>
</tr>
<tr>
<td>3</td>
<td>TC3</td>
<td>Thermochromic ink</td>
<td>Reversible</td>
<td>Commercial</td>
<td>Flexography</td>
</tr>
<tr>
<td>4</td>
<td>TC4</td>
<td>Thermochromic ink</td>
<td>Reversible</td>
<td>Commercial</td>
<td>Flexography</td>
</tr>
<tr>
<td>5</td>
<td>PC1</td>
<td>Photochromic ink</td>
<td>Reversible</td>
<td>Commercial</td>
<td>Flexography</td>
</tr>
<tr>
<td>6</td>
<td>PC2</td>
<td>Photochromic ink</td>
<td>Reversible</td>
<td>Commercial</td>
<td>Screen</td>
</tr>
<tr>
<td>7</td>
<td>PC3</td>
<td>Photochromic ink</td>
<td>Reversible</td>
<td>Developmental</td>
<td>Inkjet</td>
</tr>
<tr>
<td>8</td>
<td>PC4</td>
<td>Photochromic ink</td>
<td>Irreversible</td>
<td>Developmental</td>
<td>Inkjet</td>
</tr>
<tr>
<td>9</td>
<td>IN1</td>
<td>Oxygen indicator</td>
<td>Irreversible</td>
<td>Developmental</td>
<td>Flexography</td>
</tr>
<tr>
<td>10</td>
<td>IN2</td>
<td>Oxygen indicator</td>
<td>Irreversible</td>
<td>Developmental</td>
<td>Inkjet</td>
</tr>
</tbody>
</table>

Table 1: Different functional ink and indicator technologies used in this paper.
The relationship between product spoilage and indicator colour was not under the scope of this study. The inks are summarized in Table 1.

Surface tension of the developmental inks was measured with Aqua Pi Instrument from Kibron Inc. Viscosity was measured with Anton Paar MCR-301 rheometer at +20°C.

The following printing equipment were used for the printing trials (Figure 3) and three parallel sheets were printed for each test point:

1. Laboratory scale flexography printer with camera-based alignment: RK Flexiproof
2. Laboratory scale screen printer: EKRA
3. Inkjet printer with laboratory scale print-heads: DMP-2831 (Fujifilm Dimatix) with single-use 16 nozzle printheads and 10 pl drop size, printing resolution 1270 dpi.

The following paper and plastic based substrates were used in different trials:

1. Color copier paper 100 g/m² (Copy paper, Paper 1)
2. Coated photographic paper PDQ Gloss Photobase 250 µm thick (Photographic paper, Paper 2)
3. Adhesive label for office copier (Photographic paper, Paper 3)
4. Woodfree, multicoated silk matt art printing paper Lumisilk from Stora Enso, 130 g/m², 111 µm thick (Photographic paper, Paper 4)
5. Polyester plastic film DuPont Melinex 504, 125 µm thick, PET (PolyEthylene Terephthalate) (PET)

The substrates and with which inks they were used are summarized in Table 2.

Printing layout consisted of QR-codes or Data Matrix codes with different cell sizes: cell size 0.25 mm - 1.5 mm with 0.25 mm intervals. Multiple codes (4-8) of the same cell size were on one sample - the smaller the cell size, the more parallel codes on single sheet. For each test point five parallel samples were printed resulting in total of 20-40 codes of each cell size. The codes contained website address: http://www.vtt.fi. The choice of the code technology used in each experiment was made based on other aims of a larger research work carried out simultaneously, and the objectives of that work are outside the scope of this paper. However, both QR code and Data Matrix are technologies based on black and white squares, and the same reading devices can be used with them. Thereby, the use of different code technologies was not considered to affect the obtained results and their comparability with each other in the scope of this paper.

Figure 3: Printing machinery used: a) flexography printer, b) screen printer and c) inkjet printer.
Capabilities of the mobile phone camera and integrated code scanning software would certainly affect code readability. To avoid this issue, a mobile phone with high-quality camera was used. The printed codes were scanned with Samsung Galaxy S7 mobile phone, and for each test point 10 parallel scans were made. Decoding was done with UpCode code reader software installed on the phone that is capable of decoding both QR codes and Data Matrices. Code scanning was carried out at threshold temperature of the thermochromic inks or under UV light for photochromic inks. The indicators were scanned in conditions causing colour change, i.e. high enough humidity or oxygen concentration.

### 3 RESULTS

#### 3.1 Thermochromic inks

**3.1.1 Flexography**

Flexography printing of a reversible green thermochromic ink (TC1) that is green < +8 °C and clear > +8 °C was carried out on copy paper (Paper 1). Two different anilox rolls were used: 18 cm³/m² and 38 cm³/m². The larger the anilox volume the more ink is transferred to the substrate. More ink produces a darker printed area, but also causes more ink spreading having an effect on detail rendering. One, two or three ink layers were printed. Printed samples are presented in Figure 4. The codes with cell size 0.5 mm or more were of good print quality and cells

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Code</th>
<th>Type</th>
<th>Ink/indicators</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Paper 1</td>
<td>Copy paper</td>
<td>TC1, TC2, TC3, TC4, PC1, PC2, PC3, PC4</td>
</tr>
<tr>
<td>2</td>
<td>Paper 2</td>
<td>Photographic paper</td>
<td>TC2, PC3, PC4</td>
</tr>
<tr>
<td>3</td>
<td>Paper 3</td>
<td>Self-adhesive paper</td>
<td>TC2, PC3, PC4</td>
</tr>
<tr>
<td>4</td>
<td>Paper 4</td>
<td>Coated paper</td>
<td>IN1</td>
</tr>
<tr>
<td>5</td>
<td>PET</td>
<td>Plastic film</td>
<td>IN2</td>
</tr>
</tbody>
</table>

Table 2: Different substrates used in the experiments.

![Figure 4: Data Matrix codes with cell size 1.00 mm printed with a green thermochromic ink (TC1) when at < +8 °C.](image)
were reproduced properly, but there was too much ink spreading in codes with cell size 0.25 mm. An example of the codes with different cell size is presented in Figure 5, where the poor print quality of the smallest codes can be seen.

Color change from clear to color occurred in less than 30 seconds at <8°C. The codes were scanned when placed on top of ice bags in order to have the green color visible as a test setup. The codes were successfully decoded down to cell size 0.5 mm.

3.1.2 Inkjet

Thermochromic inkjet ink (TC2) was formulated from the pigment dispersion by mixing it with water and 1,2-propandiol. The pigment concentration was 5 wt-%. Surface active agent Dynol 604 (0.05 wt-%) was added for adjusting the surface tension. 1 wt-% of polyvinyl pyrrolidone (PVP) was used as a binder. The resulting surface tension was 33.4 nN/m and the viscosity was 4.75 cP with Newtonian behavior. Copy paper (Paper 1), photographic paper (Paper 2) and self-adhesive label paper (Paper 3) were used as printing substrates. For printing ten ink layers were required for achieving the sufficient code darkness (Figure 6).

On all three substrates, the thermochromic QR codes printed with TC2 were successfully scanned and decoded down to 0.50 mm cell size at temperature 0.25 mm, 0.50 mm, 0.75 mm and 1.00 mm printed with 38 cm3/m2 anilox and one ink layer.
above +47°C. With a smaller cell size (0.25 mm), the ink spreading caused the codes not to be decodable.

3.1.3 Combination of two functional inks

It was also evaluated if it is possible to print two thermochromic inks on a single code in order to evaluate more complex Smart Tags that can sense multiple environmental variables. For this experiment, two commercial flexography inks were used: TC3 and TC4. The first ink is red under +47°C and transparent above. The second ink is black under +31°C and transparent above. Printing was done on copy paper (Paper 1) with codes that were 2 cm, 3 cm and 4 cm squares corresponding to cell sizes 0.9 mm, 1.4 mm and 1.8 mm, respectively. It was found that printing two functional inks into a single code is a feasible solution with good enough registration (Figure 7). This idea can be utilized for making codes that contain cells that disappear and other cells that appear when exposed to certain external stimuli. However this solution is not suitable for production, as Data Matrix codes with enough identical and enough non-identical but still locationally matching cells, has its limitations and it is complex to find Data Matrixes that can be used together from encoding point-of-view.

3.2 Photochromic inks

3.2.1 Flexography

Flexography printing of a red reversible photochromic ink (PC1) was carried out on copy paper (Paper 1) with different anilox: 18 cm³/m² and 38 cm³/m². Only one ink layer was printed in all cases since it already provided dark enough printing and multiple layers spread too much. After printing, the codes are clear, and when exposed to UV or sunlight turn red in approximately 10-30 seconds. The red color starts to fade immediately when the light source is removed. Printed samples are presented in figures 8 and 9. The codes with cell size 0.5 mm or more were of good print quality and cells were reproduced properly, but there was too much ink spreading in codes with cell size 0.25 mm.

The printed codes where scanned and decoded correctly down to 0.25 mm cell size under UV light. Color change from clear to red occurred in less than 30 seconds.

3.2.2 Screen printing

Screen printing of a red reversible photochromic ink (PC2) was carried out on copy paper (Paper 1) in order to produce even darker codes than by flexography printing. Typically, screen printing produces thicker ink layers than by flexography printing.
One, two or three ink layers were printed. After printing, the codes are clear, and when exposed to UV or sunlight turn red in approximately 10-30 seconds. Printed samples are presented in Figure 10. The codes with all the cell sizes were of good print quality and cells were reproduced properly. When multiple layers were printed, the ink started to sometimes crack and de-attach from the substrate (Figure 10). Since the printing was done with a laboratory scale printer, the ink started to dry on the screen mesh quite soon. Thereby, this ink seems to be more suited for high speed printing where drying probably won’t cause any issues.

The screen printed codes were decodable down to cell size 0.25 mm under UV light. The red color started to slowly fade when the light source was removed, but remained slightly red even after 24 hours probably due to properties of the ink pigment. This might confuse users and printing process and tag layout should be designed to eliminate the effect from this residual colour change.

3.2.3 Inkjet

Reversible photochromic inkjet ink (PC3) was formulated with propyl acetate and 1,2-propanediol into a 5 wt-% pigment concentration. Reactol™ 1717E with 5 wt-% concentration was used as a binder. The resulting surface tension was 26 mN/m and viscosity was 3.5 cP with Newtonian behavior. The printability of the ink was challenging and there was much ink spreading. Thereby, good enough print quality was achieved only on photographic paper (Paper 2) (Figure 11), although also copy paper (Paper 1) and self-adhesive label paper (Paper 3) were used. However, the ink was very dark even with one ink layer on the photographic paper. The codes were decodable with the mobile phone down to 0.50 mm cell size.

Irreversible photochromic inkjet ink (PC4) was a mix of water, 1,2-propanediol and 0.05 wt-% Dynol 604. Dye concentration was 5 wt-%. The measured surface tension was 30 mN/m and viscosity 5 cP with Newtonian behavior. Ten ink layers were required for sufficient darkness. Copy paper (Paper 1), photographic paper (Paper 2) and self-adhesive label paper (Paper 3) were used as printing substrates, but on copy paper the print quality was too poor for the codes to be scanned (Figure 12). The codes were scanned and decoded on the other substrates successfully down to 0.50 mm cell size.

3.3 Indicators

3.3.1 Oxygen indicator

The oxygen indicator presented here is an ink printed directly on the packaging material or on a sticker attached inside the package to monitor the product headspace. The indicator provides irreversible evidence of leakage or tampering of a package. The indicator can be handled and stored in air at room temperature. After packaging (preferably with scavenger), the indicator is activated automatically or by touching the indicator with a heated tool. The indicator turns from yellow to green and eventually to blue when in contact with oxygen. The colour change will start immediately when in contact with oxygen and the

Figure 10: Data Matrix codes with 1.00 mm cell size screen printed with a reversible photochromic ink (PC2) when under UV light.  

Figure 11: QR code with 0.75 mm cell size printed with the reversible photochromic inkjet ink (PC3) on photographic paper (Paper 2).
reaction speed increases when the amount of oxygen increases. Thereby, the colour change is slower with small package leakage and faster with bigger leakage. This provides an opportunity to calculate correlation between package leakage and spoilage. The oxygen indicator developed for indicating leakage of vacuum and modified atmosphere packaging was based on a reduced redox dye. For ink formulation, the active components and the binder polymer were dissolved in solvents selected based on suitability for printing.

The formulated oxygen sensitive ink (IN1) was flexographically printed on PET with 38 cm$^2$/m$^2$ anilox. Since the oxygen indicator has to be placed inside a package and seen through the substrate, a transparent plastic film was used as a substrate. Five or ten ink layers were required for sufficient darkness. The codes were of good print quality and the cells were reproduced properly. The color of the print was green and the print color did not change during storage in air. The oxygen sensitive codes were packed in an inert oxygen-free atmosphere (nitrogen) together with an oxygen scavenger. The packages without other content were stored at room temperature until the next morning. The dye underwent an automatic chemical reaction during the storage in oxygen-free atmosphere at room temperature resulting in a color change to yellow. Thereby, the oxygen sensitive prints were activated. When the packages were opened, the color changed to green in a less than 3 minutes. The colour did not change from yellow to blue due to relatively thin layer of the indicator ink. However, even the colour change from yellow from green was considered to be sufficient for a mobile phone to detect this change. The color changes were documented by taking photographs. The function of the indicator is illustrated in Figure 13.

The codes printed with 10 ink layers were decodable with mobile phone down to cell size 0.5 mm. The codes printed with 5 ink layers were more challenging as the color shade was quite pale and no reliable decoding was achieved.

Figure 12: QR codes with 0.75 mm cell size printed with the irreversible photochromic inkjet ink (PC4) on a) self-adhesive label paper (Paper 3) and on b) photographic paper (Paper 2).

Figure 13: Function of the printed oxygen indicator (IN1). Cell size 1.50 mm, 10 ink layers.

Figure 14: Principle of the developed humidity indicator. The two inks are printed adjacent to each other. Upon exposure to humidity the two inks diffuse and result in a color change. The color change can be controlled by design: from yellow and blue to yellow, from yellow and blue to blue, or from yellow and blue to green.
3.3.2 Humidity indicator

The humidity indicator (IN2) presented here consisted of two inks: 1) indicator ink including a moisture absorbing substance and an indicator dye, and 2) acid ink containing an organic non-volatile acid. Moisture absorption into the system enables the diffusion and subsequent reaction of the inks resulting in a visual color change as presented in Figure 14. The speed and sensitivity of the reaction can be controlled by modifying the layout, the substrate and the amount of the inks. The colour change is reversible so after exposure to humidity the indicator does not revert back to its original state. Sensitivity, such as speed of the colour change or reactivity of even small humidity changes, can be tailored to meet specific product and monitoring requirements by modifying the ink recipe and/or the printing layout.

The indicator ink contained water, 1,2-propandiol, 0.05 wt-% Dynol, 2 wt-% PVP, calcium chloride (CaCl$_2$) for moisture adsorption, and the indicator dye. Natrium hydroxide (NaOH) was used to adjust the pH of the ink. The acid dye contained water, 1,2-propandiol, 2 wt-% PVP and lactid acid. A high quality substrate with proper pH, liquid transfer capability and humidity absorbing properties were required. Thereby, the inks were printed together on the coated paper (Paper 4): first a layer of indicator ink and then a layer of acid ink on top (Figure 15). One ink layer was enough for sufficient darkness. The resulting codes were scannable down to 0.50 mm cell size. There was too much ink spreading with 0.25 mm cell size for that to be detectable.

Finally, a principle to integrate the humidity indicator with a 2D barcode was designed. For this purpose code and sensor combination invented by UpCode Ltd. was utilized [16, 17]. The idea is that the 2D barcode contains a bar at the bottom of the code. The bar can be visible or not depending on the environmental condition it is monitoring, such as temperature or light. The code contains different meta-data depending on the state of the functional bar, and UpCode reader can detect this.
change in order to provide different information based on state of sensor. This idea was utilized by printing the acid dye (yellow) as the background of the code and the indicator dye (blue) as the bar at the bottom of the code as presented in Figure 16. The code itself was printed with a regular black ink. When the code was exposed to humidity, the bar disappeared as the indicator ink spread into the acid ink resulting in a light greenish background for the code. UpCode reader was able to detect this change in the appearance of the bar and directed to a different website (Figure 17). Thanks to the visually clear colour change from blue to yellow, no false readings were experienced.

4 DISCUSSION

This paper has evaluated the suitability of different functional ink and indicator technologies as enablers for Smart Tag services for intelligent packaging. The different ink and indicator technologies have been found technically suitable to be printed as part of Smart Tags and decodable by mobile phone reader. Of course, thorough investigations in end use conditions would be needed to verify suitability for real-life scenarios. The main technical challenges have been found among formulating functional inks for inkjet printing. Multiple ink layers are required in order to achieve sufficient darkness for the tags. There the ink-substrate compatibility is also a critical issue in order to achieve sufficient print quality for decoding.

The evaluated ink and indicator technologies have potential in multiple applications both for intelligent packaging and outside packaging area. The potential applications include:

- Offering guidance on how to use the products and recycle the packages. Guidance can be offered in interactive manners, e.g. by utilization of augmented reality (AR), online diaries and videos.
- Monitoring correct storage and usage temperature of diagnostic tests sensitive to environmental conditions.
- Indicating safe conditions for e.g. water use.
- Providing ground source tool for environmental observation.

For each application selection of the Smart Tag enablers to use have to be considered case-by-case. The selection has to be made at least among the following options:

- What parameter has to be monitored? Are suitable inks or indicator available?
- Which printing technology to use? Are unique tags required? Can the existing printing processes and equipment be utilized?
- Is a reversible or an irreversible solution required?
- Which 2D barcode technology to use? Is suitable mobile scanning software available?

This paper has targeted to address all these considerations and to prove that no matter what the application requirements are, suitable technology enablers for creating Smart Tags and services based on them exist, or can be developed. In addition, digital service building blocks and models have to be considered, but those are out of the scope of this paper. More detailed specification of ICT architecture can be found e.g. by Gligoric et al [17] and Hakola et al [23].

Sustainability as a business approach is becoming widely adopted by companies and organizations around the world and packaging sector is no exception. This means that any additional active item, including Smart Tags, have to be considered also from sustainability perspective without affecting the waste and end-of-life management processes of the packages. Here the sustainable materials are the key. Since Smart Tags described in the paper are based on inks similar to any printing ink, they should not interfere the existing recycling and composting
processed compared to e.g. RFID tags that are electronic devices with metals and chips.

Consumer perception is another important aspect to keep in mind when using Smart Tags. Consumer trust could certainly improve when more information on product history and current state is provided, meaning e.g. food quality and food safety. However, the downside might be that consumers get confused of all information provided. For example, Smart Tags indicating product not being in optimal condition, might confuse consumers and actually create mistrust. Therefore, it should be carefully evaluated which Smart Tags should be suitable for consumer use, and how they need to be visualised for consumers to avoid confusion. Still, Smart Tags can be utilised to communicate with different actors in the value chain during transport, storage, and use, and at the end of the life cycle.

5 CONCLUSIONS

Smart Tags are dynamic printed elements sensitive to environmental conditions that enable novel services for intelligent packaging. The benefits offered by Smart Tags include consumer engagement, direct contact between consumers and brand owners, product safety and quality control, and anti-counterfeiting. This paper has shown that it is possible to produce Smart Tags with different types of functional inks and printed indicators in order to create Smart Tags services.

Table 3 summarized the different experiments done with different ink and indicator technologies. Based on the experimental work carried out in this paper functional ink and indicator technologies are suitable enablers for Smart Tags. The size of individual cells can be at least 0.50 mm, sometimes even down to 0.25 mm depending on material and process combinations, and still scanned successfully by mobile phones. If the information content stored in a Smart Tag is a website address with approximately 20 characters (26 x 26 cells), this corresponds to a physical code size of 6.5 mm (0.25 mm cells) or 13 mm (0.50 mm cells). This size of a Smart Tag should be feasible to include to a package design. However, such small Smart Tags might be difficult for the end user to find. It might be worth to consider using larger cell size, thus also making code decoding in challenging conditions (e.g. low lighting level) easier.

Specifically the commercially available inks (TC1, TC2, PC1, PC2) were easily printable and decodable. Printed indicator technologies were also shown to have potential in Smart Tag applications with the same smallest cell size achieved that with the commercial inks. Since these technologies are ink based, they can be used in similar manner as functional inks for printing any layout together with 2D barcodes.

From printing perspective and based on the experiments it can be conclude that analog and digital printing are both suitable printing technologies for producing Smart Tags depending on application and end use needs. Digital printing methods are more suitable when unique Smart Tags are required, and analog printing when high-throughput without unique Smart Tags is required. However, a significant challenge with some of the inkjet printed technologies used in this paper, is the need to print multiple ink layers. This causes challenges for industrial printing process and might not be a feasible solution. Therefore, the inkjet inks still require development in order to be suitable for industrial scale Smart Tag services.
Table 3

<table>
<thead>
<tr>
<th>Ink No.</th>
<th>Type</th>
<th>Printing</th>
<th>Substrate</th>
<th>Smallest cell size decoded</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Thermochromic ink</td>
<td>Flexography, 1-3 ink layers</td>
<td>Copy paper</td>
<td>0.50 mm</td>
</tr>
<tr>
<td>2</td>
<td>Thermochromic ink</td>
<td>Inkjet, 10 ink layers</td>
<td>Copy paper, photographic paper, self-adhesive label paper</td>
<td>0.50 mm</td>
</tr>
<tr>
<td>3 +4</td>
<td>Thermochromic ink used together</td>
<td>Flexography</td>
<td>Copy paper</td>
<td>0.90 mm</td>
</tr>
<tr>
<td>5</td>
<td>Photochromic ink</td>
<td>Flexography</td>
<td>Copy paper</td>
<td>0.25 mm</td>
</tr>
<tr>
<td>6</td>
<td>Photochromic ink</td>
<td>Screen, 1-3 ink layers</td>
<td>Copy paper</td>
<td>0.25 mm</td>
</tr>
<tr>
<td>7</td>
<td>Photochromic ink</td>
<td>Inkjet</td>
<td>Photographic paper</td>
<td>0.50 mm</td>
</tr>
<tr>
<td>8</td>
<td>Photochromic ink</td>
<td>Inkjet, 10 ink layers</td>
<td>Photographic paper, self-adhesive label paper</td>
<td>0.50 mm</td>
</tr>
<tr>
<td>9</td>
<td>Oxygen indicator</td>
<td>Flexography, 10 ink layers</td>
<td>PET</td>
<td>0.50 mm</td>
</tr>
<tr>
<td>10</td>
<td>Humidity indicator</td>
<td>Inkjet, two ink concept</td>
<td>Coated paper</td>
<td>0.50 mm</td>
</tr>
</tbody>
</table>
REFERENCES


