Bonding of Flexible Mulitlayer Printed Systems based on PET-Substrates – An Investigation of Challenges and Promising Approaches

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***Abstract* — Flexible, thin and smart multilayer printed systems have a wide range of applications, reaching from human-centred alarm systems to industrial applications like condition monitoring within the scope of Industry 4.0. During the production process for flexible mulitlayer printed systems, the bonding of the individual substrate layers is an important step. Thereby, the functionality of the components and lines printed onto the different layers must not be affected. In this paper, the challenges of the bonding process of flexible printed systems based on Polyethylene Terephthalate (PET) substrates are demonstrated and possible approaches for bonding are introduced and compared to each other.**

Keywords – multilayer printed systems; bonding of PET; transmission laserwelding; direct thermal bonding; adhesive bonding; surface activation.

I. INTRODUCITON TO PRINTED SYSTEMS

A. Potentials of flexible multilayer printed systems

The term of printed systems is used in the context of this paper for systems based upon thin polymer foils, called substrates, comprising printed components and conductive lines, in the following aggregated with the term “printed elements”. The ongoing development of functionalized inks opens the possibility to print a large variety of different materials onto varying substrates. Thereby, printing techniques are applied, that are being well established for the printing of color on paper, reaching from screen printing to inkjet printing [1].

One motivation for producing printed electronic systems is cost optimization. Using roll-to-roll techniques, the production of large quantities of printed materials is possible. Alternatively, existing products can be replaced by cheaper and therefore disposable printed systems [2]. Thereby, mainly single-layer systems are used.

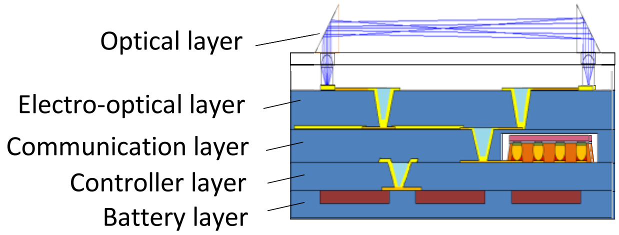

Figure 1. Visualization of the setup of a multilayer printed system by the example of an electro-optical sensor (cp. [3]).

Here, another approach is taken [3]. Printing technologies offer the potential of producing multifunctional, highly integrated smart systems. The potential can be realized by printing task-specific elements onto one side of each substrate using different printing techniques. These single layers can contain electrical circuits, sensor elements, optical or fluidic components. One approach is to separate functions between layers (see Figure 1). In order to combine the layers and to enable interaction between the different domains, the single layers are bonded, resulting in a multilayer system. The use of flexible substrates allows for the production of flexible systems.

Potential applications are diverse. When implementing sensors and wireless communication they reach from human-centered to industrial applications. Examples for human-centered alarm systems are illustrated in Figure 2. They include thin and nearly invisible smoke detectors, humidity monitoring on walls for mildew prevention or within smart plasters to prevent dehydration, e.g., of elderly people. Industrial applications of smart multilayer printed systems are conceivable for industry 4.0 and decentralized production, where condition monitoring of workpieces, machines and goods during manufacturing, transport and application is necessary [4]. Here, printing technologies can be used for generative production technology with tailor-made customized monitoring systems, allowing for small production quantities down to a single piece.

B. Setup and manufacturing of multilayer printed systems

For the setup of the printed systems described here, transparent flexible polyethylene terephthalate (PET)

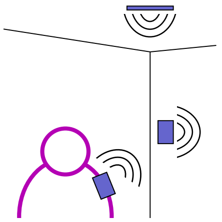

Figure 2. Visualization of examples for human-centered alarm systems: dehydration sensors or other smart plasters, smoke detectors, humidity sensors for mildew prevention.

substrates are used. In order to demonstrate the manufacturing feasibility, sheets of 150mm x 150mm are being processed, whereby the transfer to mass production techniques is considered. Conductive lines are printed on top of these substrates by using inks filled with metal particles, e.g., silver nano particles [5]. To print resistors, highly resistive inks containing, e.g., carbon particles are applied. Subsequently, the printed elements have to be sintered in order to evaporate the solvent of the ink. The particles get in contact to each other, going from a percolation-based conduction to forming of sinter necks and grain growth, ultimately leading to porous, metallic structure. Thermal sintering of silver nanoparticles occurs at temperatures of 120°C to 150°C [6][7]. Investigated alternatives are flash light sintering or current sintering.

In order to realize capacitors and field effect transistors, a stack of different materials has to be printed. Therefore, special inks have to be developed to realize the dielectric and the electrolyte [7][8]. For the time being, the printed components require large areas compared to conventionally manufactured surface mounted devices (SMD). But they are very thin and flat and thereby flexible. In order to setup fully printed electronic circuits, the integration and interconnection of these printed components on one layer is necessary. This class of integration technique is called intralayer-integration [3]. It also includes the embedding of conventional components like silicon bare dies (see Figure 1).

The second class is called interlayer-integration. It comprises the mechanical bonding of the layers as well as establishing electrical, optical and fluidic interconnections between the layers.

Two approaches of the design for electrical vias are illustrated in Figure 3. The first approach is to print a full metal pad on the lower layer and a metal frame pad placed around a hole on the top layer. By filling the hole with conductive adhesive, the two metal pads are connected electrically. Disadvantageous are thereby the change of production techniques and the protruding of conductive adhesive over the level of the top layer. Bridging steep-walled holes with conductive ink is not possible due to the evaporation of the solvent during sintering, that results in a strong reduction of ink volume. In order to enable a continuous printing process and to avoid the use of conductive adhesive, it is necessary create conical holes. The metal area on the lower layer can now be electrically connected by printing a continuous conductive line on the top layer (see Figure 3 right). The realization of conical holes, however, is more elaborate than the one of steep-walled holes.

For the production of multilayer printed systems, two general processes are being investigated:

1. Printing and assembly of all elements on the substrates and subsequent bonding of the printed substrates
2. Succession of printing and assembly of elements on one layer, bonding of the subsequent layer and printing of elements on the top layer of the bonded stack

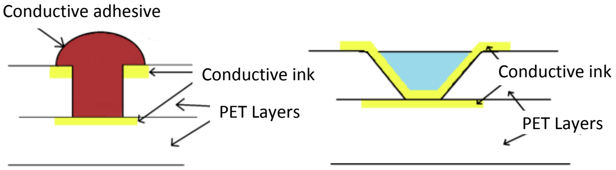

Figure 3. Design of electric vias to interconnect two layers of printed systems by filling a hole with conductive ink (left) or overprinting a conical hole (right).

Both processes are illustrated in Figure 4. The advantage of the first approach is the separation of production processes. Process parameters of printing techniques for different domains, like high sintering temperatures, do not affect the other layers. Theoretically, even the bonding of all layers in one step would be conceivable. The integration of vias, however, will require a layer-by-layer bonding process. Furthermore, the bonding of layers with assembled protruding components is hard to realize, since the bonding processes require a compression of the individual layers. This can be easier realized when using the second approach. The latter does also allow for both, printed and conductive adhesive vias, to be applied.

In Section I, multilayer printed systems and the general processes for their production were introduced. In Section II,

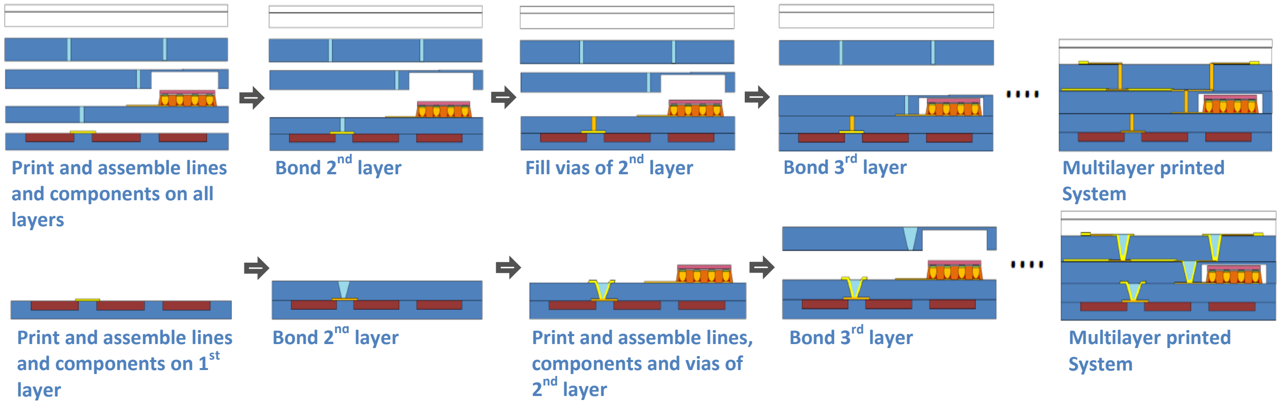

Figure 4. Investigated general production processes of multilayer printed systems: Printing and assembling of all layers and subsequent bonding (top) and bonding of empty substrates and printed layer(s) and subsequent printing and assembly (bottom).

the requirements for the bonding process to join individual printed system layers are defined, a general approach for bonding is explained and the three favored bonding methods are presented. These are transmission laserwelding, adhesive bonding and direct thermal bonding. After the comparison of the bonding methods in Section III, the conclusion of this paper is drawn in Section IV.

II. BONDING METHODS

A. *Requirements for the bonding process*

The objective of the bonding is the provision of a strong bond between the layers that allows for flexing of the final system without delamination. Thereby, the functionality of the printed system must not be affected by the bonding process. The flexibility of the bonded layers should be comparable to the flexibility of bulk material of equivalent thickness. In order to enable the second bonding process, the printability of the surface must be preserved. Appealing bond quality, including smooth surfaces, is also aspired for the final systems. The preservation of transparency is desirable but not necessary, since optical elements will presumably be omitted during bonding. The bonding time is also considered, including all process steps that are necessary to bond the substrates, e.g., application of adjuvant bonding materials. The importance of the bonding time strongly depends on the batch size of the respective system. Further aspects are bonding costs and process complexity.

For the investigation, PET Melinex ST 506 substrates of 125 µm or 175 µm thickness are being used to ensure sufficient stiffness for substrate handling [9]. In future work, it is envisaged to reduce substrate thickness to enhance the flexibility of the layer stack. The substrates have a glass transition temperature of 78˚C [10] and a melting point of 265˚C [9]. Due to the roll–to-roll manufacturing process, the thermal shrinkage of the substrate is anisotropic.

In experiments with the different bonding techniques, first the parameters have to be determined that enable a sufficiently strong bond between the layers. The bond strength will be analyzed using T-peel tests in accordance to DIN EN ISO 11339. Afterwards, the possible damage of the bonding process on the printed elements is investigated for the given parameters. In order to determine the influence on conductive lines, for example, special standardized test structures were designed (see Figure 5). Their conductivity is measured before and after bonding.

B. *General approach*

The investigated bonding techniques can be classified as direct bonding and indirect bonding [11]. Adhesive bonding, using either liquid adhesives or lamination films, is a well established indirect bonding technique. For direct bonding, on the other hand, no additional material is added at the interface. One approach is to transfer thermal energy into the bond area – either localized or simultaneously over at the entire surface. Another possibility is to influence the polymer surface. Hereby, either solvent bonding techniques or different kinds of surface treatment and modification can be used.

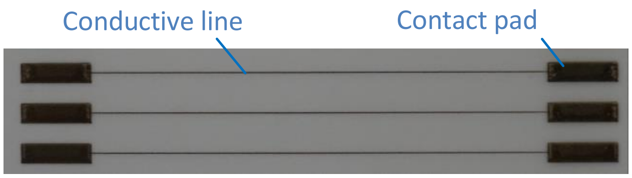

Figure 5. Test structures of printed conductive lines.

Due to the material and geometry of the given application, the three approaches that are further investigated and compared to each other are laserwelding, direct thermal bonding, if necessary in combination with surface activation, processes and adhesive bonding.

In order to reduce bonding complexity and bonding costs, the aspired objective is to find a layout-independent bonding process, that can be equally applied to any printed system. If a negative impact of the bonding process on the printed structures cannot be avoided, however, a potential compromise solution is to omit the printed structures during the bonding process. The resulting setup is illustrated in Figure 6. The most important areas to be bonded are the outer substrate frame and the area around vias to increase via stability. Additional bonding areas must be defined in order to achieve the aspired bonding strength. The individual areas are staggered to avoid negative effects of stacking, like local stiffening or interferences of the bonding process with already bonded interconnections. It is obvious, that this approach will not only result in a loss of available element area, but also in extensive effort. Special bonding keep-out areas have to be included into the circuit design rules of the printed systems, and individual programming of bonding equipment is required for every layout. The flexibility, on the other hand, might be increased if only small areas of the substrates are bonded. The effects of the application of layout-dependent as well as layout-independent bonding will be analyzed for every investigated bonding technique.

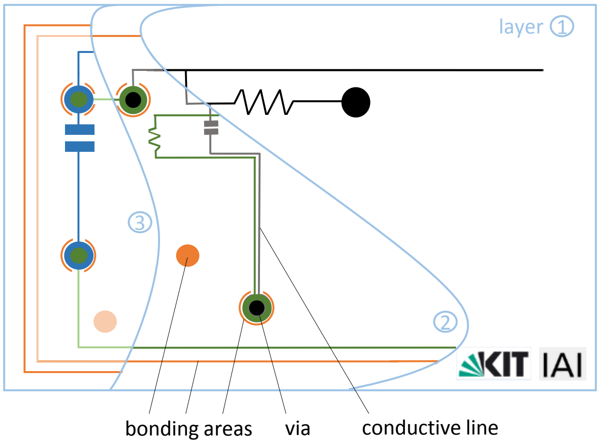

Figure 6. Illustration of layout-dependent bonding procedure.

C. *Transmission laserwelding*

Transmission laserwelding of two polymer parts is based upon heat generation in the joint area of the parts that are tightly clamped together [12][13]. Therefore, the laser light passes the upper part that is transparent for the wavelength of the laser. In the joint area, the laser light is absorbed and the absorbed energy results in heat generation. This leads to melting of the polymer surfaces and thus to bonding of the clamped parts.

One approach to realize absorption of laser light in the joint area is to use lower bond parts that are absorbent for the laser light [13]. In order to weld two transparent polymer parts, special coatings like a thin sputtered carbon layer or Clearweld™ are applied at the bond interface [14][15]. Clearweld™ is a liquid that, once applied on a foil, quickly dries to a thin layer absorbing in the wavelength range of 900 nm to 1100 nm. The use of fiber lasers with higher wavelength than infrared even allows laserwelding without additional coatings for a variety of thermoplastics [16]. Since the bond is formed between the molten surfaces of the thermoplastic polymers, the achievable bond strength is very high, up to the cohesive strength of the bulk material. The local transfer of heat leads to thermal stress and can result in deformation of the polymer parts.

For the bonding of transparent PET foils that shall be used for the production of transparent multilayer systems, Clearweld™ is chosen as absorbing interface layer. The Clearweld™ can be easily applied by printing techniques or doctor blading, resulting in a thin uniform layer on the PET surface. During the laserwelding process, the greenish-yellow Clearweld™ layer is consumed and the final bond is again transparent [12]. Both, the Clearweld™ application and the subsequent laserwelding process, can be applied either localized for layout-dependent design or over the entire area of the layers. For layout-dependent localized application of the Clearweld™, either masks, structured rolls or individually programmed printing processes can be applied. Similarly, the laserwelding can be applied layout-dependent using masks or individual laser programming.

In order to protect the printed elements, the Clearweld™ is applied to the lower, blank side of the top layer. Using the first general production process described in section I.A, the Clearweld™ is either applied before or after printing and assembly of all elements on one layer. If applied before printing, the Clearweld™ is exposed to heat and mechanical forces caused by sintering and handling processes that might damage the coating. During a Clearweld™ application after printing and assembly, on the other hand, the printed elements are exposed to handling forces. Protruding elements would impede a conformal coating. Thus, the implementation of the second process is probably more advantageous. The coating is applied to an empty substrate, laserwelded to a lower printed layer and subsequently printed on and assembled. The main challenge is to preserve a printable surface by preventing deformation due to localized thermal stress intrusion.

In first experiments depicted in Figure 7, the applicability of laserwelding on transparent PET substrates coated by Clearweld was demonstrated using a laserwelding machine built within the framework of the SMARTLAM project [17]. The aim of the current investigations is the definition of laserwelding parameters that enable suitable bond strength and do not affect the printed elements.

D. Adhesive bonding

For adhesive bonding, an intermediate layer of adhesive is applied to the surface of at least one bonding part. The bonding parts are then pressed together and the adhesive is cured. The associated reaction of linking the single monomers of the adhesive to form large molecules is called polymerization. Adhesives can be classified in accordance with the polymerization mechanism: polyaddition or chain reaction polymerisation and polycondensation or step reaction polymerization [18]. Curing is enabled or supported by processing conditions like elevated temperature, pressure or UV-irradiation [19]. Another approach is the use of two different components that start curing after being mixed together. Adhesive bonding can be used to bond similar or dissimilar bonding parts. If choosing a suitable adhesive matching with the bonding parts, very high bond strengths can be realized [18][19]. Though adhesive bonding is a well-established bonding method, the application of uniform layers of adhesive on large areas is challenging. The introduction of an additional material into a system might reduce the system stability, e.g., to temperatures and can be critical for medical applications. For optical applications, special gradient-index matched adhesives can be applied. One advantage of adhesive bonding is that the influence of uneven surfaces, e.g., due to protruding elements, can be compensated at a certain extent.

There is a variety of adhesives designed for PET bonding (e.g. [20][21]). Using adhesive bonding for the large area substrates, the adhesive can be applied in form of a liquid film directly before the bonding process. Alternatively, the use of precoated PET foils containing a dry film of adhesive is possible. Methods of adhesive application are dispensing, doctor blading, dipping, spray-coating etc. or the use of adhesive films. Surface treatment can be applied in order to increase the wettability of the PET surface and thus the molecular forces between adhesive and substrate, leading to improved bond strength [18][22][23].

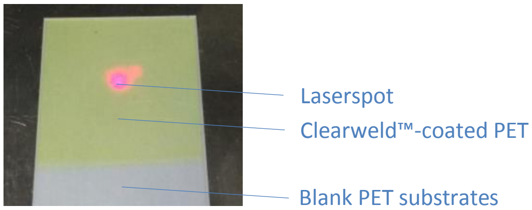

Figure 7. Laserwelding of Clearweld™-coated PET substrates.

A possible impairment of the printed elements can be caused either by chemical reactions between printed elements and adhesive or supporting conditions like UV-irradiation, or by mechanical stress during adhesive application. The latter can be omitted by the application of the adhesive only onto the lower, blank side of the top substrate or by using sensitive methods of application. The occurrence of chemical reactions has to be investigated for all types of ink being used for the production of a printed system, or has to be avoided by applying layout-dependent adhesive bonding techniques. Another aspect to be considered is that vias punched into the top substrate must not be filled by liquid adhesive. For layout-independent application, this can only be avoided by using pre-coated PET substrates allowing for punching via holes into substrate and coating. The lowest substrate is thereby not coated. When using pre-coated substrates for the first production process, special care must be taken during handling and preceding production steps in order not to damage the adhesive layer or to prematurely activate curing.

E. *Direct thermal bonding*

During direct thermal bonding, the two bond parts are heated and simultaneously pressed together. Usually, heated rolls are used to enable a high surface pressure of the bond area (see Figure 8). The combination of pressure and temperatures near or above the glass transition temperature leads to polymer softening in the bond area, enabling interdiffusion of polymer chains [11]. The achieved bond strength is very high, ideally reaching up to the cohesive strength of the bulk material. The required high temperatures, however, may cause deformation.

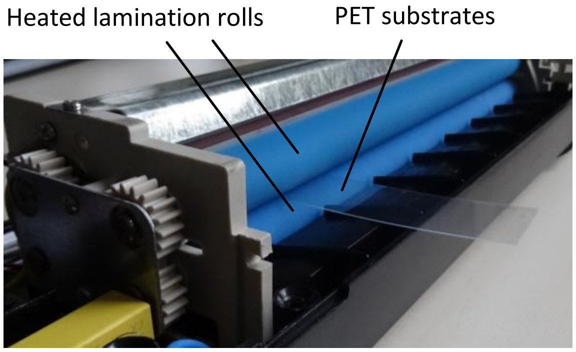

Figure 8 Direct thermal bonding of PET substrates

When using the relatively simple process of thermal bonding in order to bond thin PET substrates, all elements are heated. A layout-dependent protection of individual areas during the bonding process is not possible. One approach to reduce the process temperature significantly is to activate the substrate surface before bonding. Surface activation methods include, amongst others, the use of coupling agents [24], ultraviolet, ozone and plasma surface treatment [25][26]. Depending on the activation procedure, a layout-dependent masking of the printed structures is conceivable during this process step.

For both general bonding processes, all printed elements and mounted discrete components must withstand the bonding temperature and pressure, excluding the topmost layer, when using the second approach. Anisotropic thermal shrinkage and subsequent bending of the substrate must be prevented. The approach of first printing and assembling the individual layers, additionally leads to mechanical stress of the elements applied directly by the lamination roll. A roll-based thermal bonding of layers containing protruding components is not possible.

Due to the simplicity of the process, the thermal bonding will be further investigated, presumably in combination with surface activation methods. For first experiments, a thermal lamination machine (Dahle 70453) is used.

III. COMPARISON AND DISCUSSION

In Table 1, an overview of the three different bonding techniques that were described in the above sections is presented. Based upon the literature review and first experiments, estimates about the fulfillment of the main requirements of the bonding techniques are made. Thereby, some of the assumptions have to be further investigated in practical tests.

With transmission laserwelding, very high bond strength can be achieved. This is reduced if only parts of the area are welded, using the layout-dependent approach. The bond quality has to be investigated, especially regarding the printability on the top surface. Due to the serial process of local heating, visible lines or patterns resulting from the scanning of the Laser beam have to be expected and local deformation might occur, leading to uneven surfaces or bending of the entire substrate. The Clearweld™ coating process step and the serial welding result in rather slow bonding time. The process complexity is low to medium. Layout-dependent programming of the welding machine or even localized coating increase complexity. The costs of Clearweld™ coating and laser machines are medium, extra programming time and/or the production of masks increase

TABLE I. COMPARISON OF DIFFERENT BONDING TECHNIQUES TO BE APPLIED FOR BONDING FLEXIBLE TRANSPARENT PET SUBSTRATES CONTAINING PRINTED ELEMENTS TO SETUP MULTILAYER PRINTED SYSTEMS. ASSESMENTS OF THE DETERMINING FACTOR, REACHING FROM ++ AS VERY GOOD TO – FOR VERY BAD PERFORMANCE.

Bonding technique		**Bond strength**	**Bonding quality**	**Bonding time**	**Process complexity**	**Bonding costs**	**Flexibility of the system**	**Trans-mission**
Laser welding	Layout-independent	++	0 to +	-	0 to +	0	0	-- to -
	Layout-dependent	0 to +	- to +	-	- to 0	- to 0	0 to +	- to 0
Adhesive bonding	Layout-independent	+ to ++	+	-	0 to +	- to +	--	-- to 0
	Layout-dependent	0 to +	-- to 0	-	- to 0	- to 0	-	- to 0
Direct thermal bonding	Without surface activation	0 to +	- to +	++	++	++	0	- to 0
	With surface activation	+ to ++	+ to ++	+	+	0 to +	0	- to 0

the costs. The flexibility of the system is not affected by the laserwelding process, the reduction of the transmission caused by visible lines or unused Clearweld™ coating has to be further investigated. Due to the high laser power that has to be introduced in order to melt the PET surfaces, an impairment of the printed elements must be expected in the bonded areas.

Choosing the appropriate adhesive, very high bond strength can be realized by adhesive bonding. The reduction of bonded area in the layout-dependent approach reduces the overall strength. Applying the adhesive onto the entire area, good results can be achieved for the bond quality. Depending on the adhesion layer thickness, existing unevenness of the substrate surfaces might even be compensated. For layout-dependent adhesive bonding, on the other hand, uneven surfaces have to be expected due to the local intrusion of an extra layer. The overall bond time, including the application of adhesive, possibly with preceding surface activation, bonding and curing is rather high. The complexity and thus the bonding costs strongly depend on the need of surface activation, choice of adhesive and method of adhesive application and curing. They are increased for layout-dependent bonding due to programming or the use of masks. The flexibility is reduced due to the additional layer. The impairment of transmission depends on the choice of the adhesive. Possible damage of the printed elements due to chemical reactions of the ink with the adhesive have to be further investigated.

Using direct thermal bonding, ideally very high bond strengths can be realized. Presumably, a preceding surface activation is necessary to achieve the desired bond strength and/or to reduce the process temperature. If thermal stresses can be avoided, the bond quality is expected to be good. The fast bonding process and the low process complexity result in relatively low bonding costs. Time, complexity and costs are significantly increased if the implementation of the additional processing step for surface activation is necessary. The flexibility of the system will not be affected by the bonding method. Minor or no interference of the transmission is expected. At temperatures of 120°C or more, substantial influences of the bonding process on the substrate and the printed elements have to be expected.

IV. CONCLUSION

In this paper, three selected bonding techniques were described that can be applied for bonding of transparent flexible PET foil substrates containing printed elements to produce multilayer printed systems. The techniques are transmission laserwelding of Clearweld™-coated substrates, adhesive bonding and direct thermal bonding, presumably in combination with a surface activation process. It was shown that for all investigated bonding processes the approach of bonding a blank PET substrate on top of a printed layer or a stack of layers and subsequently printing the next layer on the blank substrate is preferable in comparison to printing all layers and subsequently bonding them. This conclusion leads to the requirement of preserving the printability of the surface of the bonded empty substrate during the bonding process. A comparison of the different bonding techniques shows, that all of them have the potential to be used for the present application if the appropriate bonding parameters can be found, enabling a strong bond and simultaneously keeping the functionality of the printed elements and the printability of the top surface. The current state of the investigation indicates that direct thermal bonding is the most promising approach due to its low complexity and relatively fast bonding time. The final decision, however, requires a comprehensive experimental investigation of the application of the different bonding techniques.

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