

DISPLAYS AND FLEXIBLE ELECTRONICS: AN EMERGING MARKET FOR ADVANCED PSAs

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One of the largest uses of PSAs in the world today is in the manufacture of displays, for everything from televisions and computer monitors to cell phones and small displays on appliances. Obviously, tape is used in the assembly of the cases and components of these devices, but this paper will focus on the use of PSAs in the construction of the display module itself.

The display market today consists largely of flat panel, thin film transistor (TFT) displays and has seen rapid growth over recent years. The construction of a flat panel display consists of many components, starting with a back light unit (BLU) that produces white light. That light is then filtered and directed through various layers of filters and prisms, before passing through an assembly containing polarizers and a liquid crystal (LC) layer, which ultimately produces the image. The most critical use of PSAs is in the bonding of polarizer films to indium tin oxide (ITO) coated glass. This assembly is shown in Figure 1 below¹.

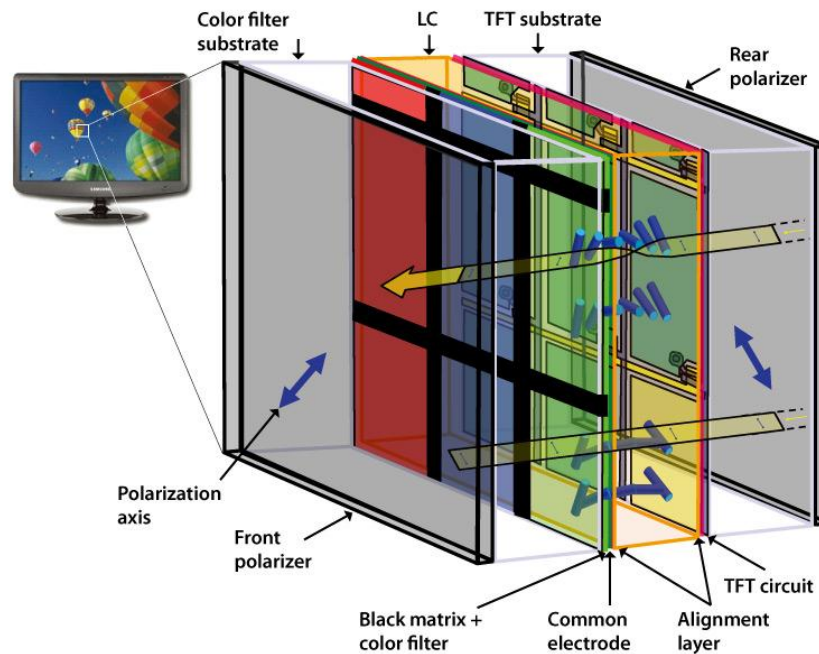


Figure 1. Polarizer films in a display.

The market size for these PSA-coated polarizer films is estimated to be about \$6.3 billion in 2012², and the recent growth is clearly seen in Figure 2. This growth comes partly from the replacement of obsolete display formats and also, of course, from the new

applications that thin LC-TFT displays helped to create. The adhesive opportunity is obviously only a small percentage of this number, but it is still an extremely significant market.

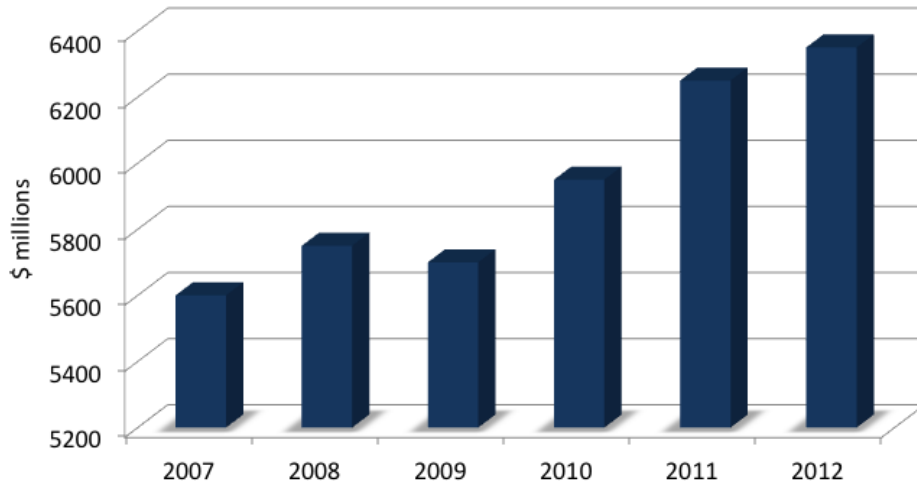


Figure 2. Market value of adhesive-coated polarizer films.

Polarizer Construction

The polarizer films are made of polyvinyl alcohol, which is treated with dichroic dyes and then stretched to orient the molecules and therefore provide the polarization effect. The PVA layer is covered on either side with a protective layer of tri-acetyl cellulose (TAC) and a PSA is coated to one of those TAC layers, as shown in Figure 3. This PSA is then used to bond the polarizer structure to the ITO glass during the display assembly.

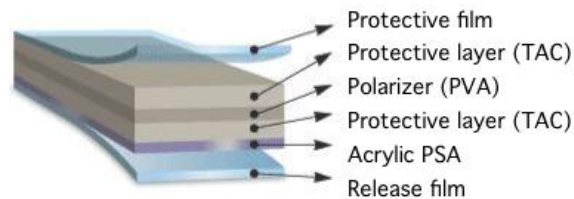


Figure 3. Polarizer film construction

PSA Requirements

The PSA used to bond the polarizer to the glass has to meet several key criteria. It must bond well to the glass, and stay in place in hot and humid environments. It must have suitable optical transparency, as the light used to create the image on the display will be transmitted through the PSA. Most importantly, it must control the stresses in the PVA so as to not cause light leakage and other artifacts in the display, which are known in the

industry as “mura” and will be explained further. Finally, due to the relatively high cost of ITO coated glass, manufacturers would like the polarizer film to be removable for up to 24 hours after lamination. This allows removal of the polarizer from the glass should lamination or coating defects be noticed, and then allows the ITO glass to be reused. Therefore, the adhesive needs to have a relatively low peel and clean removability for the first 24 hours, but build to a strong permanent adhesion over time.

Mura

Mura (斑 or ムラ) is a Japanese term for unevenness or inconsistency. In the display industry it has become associated with the description of any blemishes on a display caused by variations in luminosity. The effect is well known to anyone who has looked at a large television screen with a black image, and seen areas (often at the edges and corners) that appear lighter in color than the uniform black across the center of the screen.

There are many possible causes of mura – impurities in the crystal matrix in an LCD, flaws in the LCD cells, stresses induced in the panel during assembly, and an uneven distribution of the output from the backlight unit. Perhaps one of the most common causes is from stresses in the PVA polarizer resulting in light aberrations at the edges of the display³. The PSA used to bond the polarizer plays a key part in distributing these stresses, and minimizing any effect on the image. Such control, however, gets increasingly difficult as the display increases in size.

PSAs in use today

The merchant adhesives used in these applications today all have broadly similar physical and chemical characteristics. They are all acrylic polymers in solution, of relatively high molecular weight (and therefore relatively low solids levels) and designed to have the optical and adhesion properties needed. They are also multi-component adhesives – using a crosslinker as well as several adhesion modifiers and promoters, which must be formulated by the coater in precise ratios. In fact, these customized formulations often result in up to five component systems. The crosslinking process (commonly an isocyanate) is thermally activated and thermodynamically slow, taking up to seven days to fully complete in many cases.

Whilst the final performance in use of these adhesives is generally excellent, the operation and handling during the coating process and the display manufacture seems less than ideal. During the PSA coating process, multi-component mixing increases the opportunity for error and low solids levels lead to high energy costs for solvent removal.

Lamination of the polarizer to the ITO glass typically occurs very soon after coating (sometimes in the same location). The slow thermal cure of the adhesive therefore leads to large inventories of polarizer on glass being held until cure is complete, and the

manufacture can be satisfied with the product quality. This also slows the display assembly process.

The first parts of the manufacturing process, the making and adhesive coating of the polarizer, are shown schematically in Figure 4.

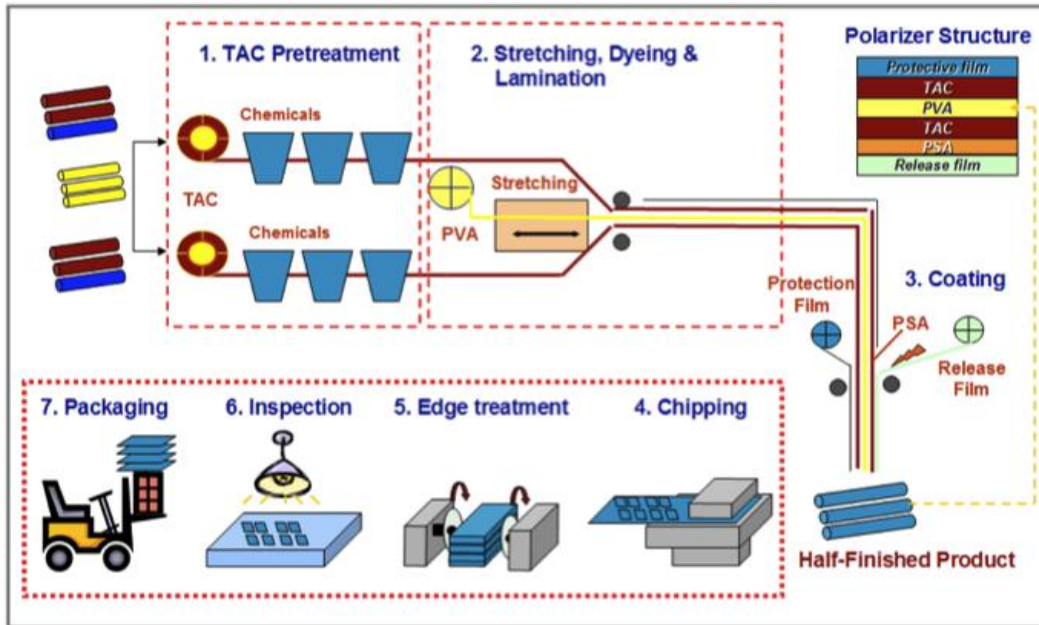


Figure 4. Polarizer manufacturing process.

A new type of PSA for displays

The process described above is not particularly efficient and yet occurs at very large scale, and is increasing every year. Display volume is growing and demands for lower cost modules are increasing alongside. This led us to ask the question: could we design a new PSA for polarizer bonding that would provide the opportunity to reshape and improve the current process? We believe the answer is yes.

We focused on addressing what we saw as the key deficiencies of the incumbent solution acrylic adhesives, namely:

- Low solids
- Multi-component mixing at the coater
- Slow cure

At the same time, it was critical to maintain the optical and mura performance, as well as the initial removability of the adhesive. We felt we could do this, whilst also creating a single component, higher solids adhesive with faster curing chemistry.

The use of an acrylic polymer was key, due to the durability and lifetime that the product would be required to have. We chose to create an acrylic polymer that would draw on our experience in making high clarity products for the window and safety film industries,

where control of optical properties is important. We also planned to use our proprietary UV technology to enable a rapid curing process⁴. Our polymer design consisted of four key components:

1. *PSA backbone monomers*: common to most PSAs and responsible for providing general PSA properties. These were also chosen to give appropriate clarity and refractive index.
2. *UV active species*: polymer-bound non-fragmenting photoinitiators and suitable reactive sites, for rapid crosslinking.
3. *Specialty monomers* to improve interaction with the TAC film.
4. *Silane functional monomers*, to improve both humidity resistance as well as bond strength to the ITO glass.

As already mentioned, the products in use today are high molecular weight (greater than 1 million), low solids solution acrylics. In choosing to create a higher solids polymer, we obviously had to reduce the molecular weight considerably, in order to still create an adhesive that could be coated with the precision and quality required. This presented some key challenges in order to still provide the mechanical properties necessary in the application. We did, however, succeed in producing a PSA with appropriate adhesion, optical and mechanical properties, using the lower molecular weight, one-part UV curing approach. We employed one of our proprietary polymer-bound photoinitiators, which have been reported on separately. The use of monomers containing trimethoxy silane functionality also helps build strong bonds to the ITO glass.

The choice of using a solution based polymer system in a UV curing application may seem unusual. Commonly, UV-curable PSAs are used today in 100% solids systems. Whilst we did consider such an approach, the quality of coating required in these applications is beyond what can be achieved with a hot melt adhesive. Coating from solution is the only viable method to produce the even, flawless coatings demanded by the display industry. Adding UV curing equipment to the end of the drying ovens is not common, but did allow us to create a system with instant cure, which provides a significant operational advantage over incumbent technology.

As the rheology in Figure 5 below shows, the polymer is a relatively high modulus material, compared to many PSAs. That high modulus creates a slow wet out, which helps with the need for short-term removability, but ultimately produces an extremely strong and durable bond to the glass substrate.

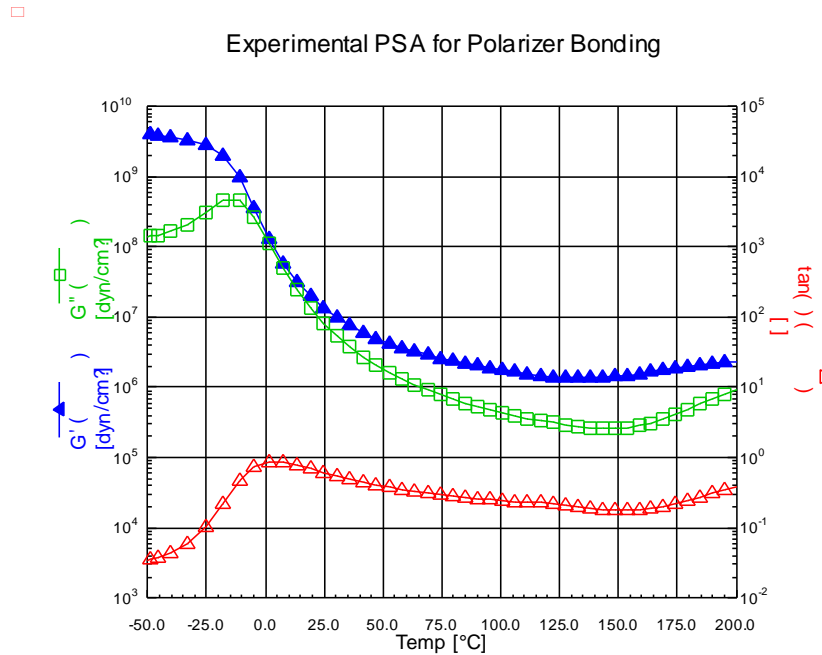


Figure 5. Rheology of polarizer adhesive.

Designing a PSA to meet the adhesion needs alone is not overly challenging – it is the optical requirements that are more complex to meet. Particularly gratifying, therefore, was the mura performance of our adhesive, which as shown in Figure 6 below, and resulted in less than 1% light leakage under industry standard tests, which was noticeably better than the reference samples, which showed up to 2.5% light leakage.

To perform the mura tests, the adhesive coated polarizer is bonded to a piece of ITO glass and placed on a back light module from a display. Although we have employed software to analyze the light leakage, many manufacturers only employ a visual comparison by a trained operator.

In summary, for conventional LC-TFT displays, we had created an innovative adhesive, which offered several in-use benefits, and had the ability to create advantage for the coater in this increasingly cost sensitive market. It offered the optical and adhesive properties necessary for the application, along with higher solids, fast UV curing, and the ease of use of a single component adhesive. Operation efficiencies at the coater can be improved, and inventories of finished coats awaiting completion of the slow thermal cure can be eliminated.

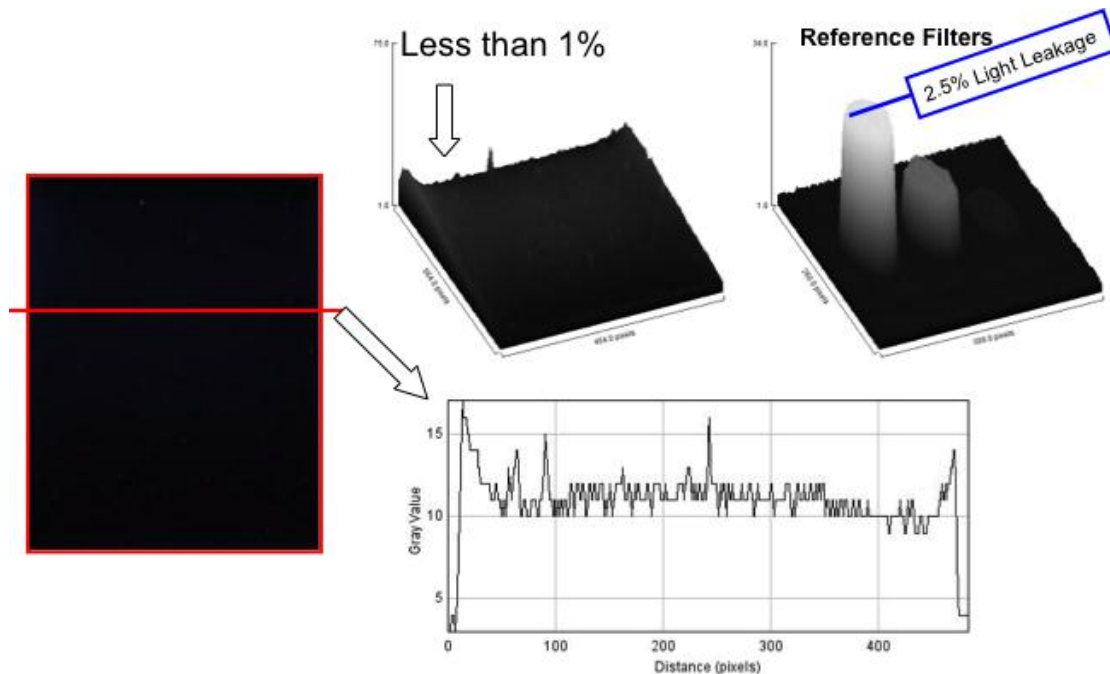


Figure 6. Mura test results.

Printed and Flexible displays – the future generation

All of the work described above represents an attempt to enter an already largely mature market space, with a disruptive adhesive development. Having started to explore the opportunities for PSAs in the display industry, we decided to broaden our scope and begin understanding the future direction of the industry and explore how we could create PSAs to meet these future needs.

The technology in use in displays is changing. Although LC-TFT displays will occupy a dominant position for many years to come, emerging technologies using OLEDs (organic light emitting diodes) are gaining increasing attention.

OLED displays differ fundamentally from LC-TFT displays in how they create the image and also offer a number of key operational advantages. All LCD-TFT displays use a backlight (either a cold cathode, or more recently an LED source) to generate light, which is then filtered through an LCD matrix to produce the image. The chemistry and material science of these processes is beyond the scope of this paper, but such assemblies result in great inefficiency in the use of the backlight, as well as traditionally limited color gamut, viewing angle and more. While advances have been made in many of the aesthetic properties of LCD-TFT displays, the inefficiency remains.¹

An OLED display dispenses with the backlight and instead uses a matrix of OLEDs across the entire viewing area, each one being a point source of light. This leads to much improved power efficiency (a black area on the screen has the OLEDs turned off –

something that never happens with the backlight on a TFT-LCD display). Viewing angle, response time and other image attributes are also improved with OLEDs, but perhaps the most far reaching advantage is the ability of OLEDs to be printed, roll-to-roll, at very low cost and on thin flexible carriers.

The projected market for printed OLED technology is extremely large, estimated to be as high as \$19 billion by 2019 in displays and lighting and \$17 billion in organic photovoltaic applications⁵. Some of this volume is a direct replacement for current display technology, but there is also growth in to new applications made possible by the thinner, lighter form factor.

Technology needs

To date, a small number of OLED displays exist in commercial applications, mostly on small sized displays – and none of them are on flexible substrates, or produced in roll-to-roll processes. Instead, they are all batch manufactured on glass carriers. This is in part due to some of the technical challenges that exist in producing a flexible display, whether made in a roll-to-roll or sheet process. This paper will focus on two areas where adhesive technology can help to solve some of the problems – in particular chip embedding and barrier properties.

In order to develop both adhesives and understanding in the field of OLEDs for flexible displays (and lighting), Henkel joined the Holst Center in 2011, an open innovation consortium based in the Netherlands that is focused on creating technologies for wireless autonomous sensors and flexible electronics⁶. The work we have done in these areas has been in close collaboration with Holst.

Chip Embedding

In order to create flexible OLED displays, it becomes important to consider the integrity of any circuitry around the edge of the display, which would also need to be flexible yet not risk any unwanted electrical contacts which could cause failure. Such circuitry would be built up in layers on a flexible carrier, to create a part similar to that shown in Figure 7.

One way of solving this potential problem is to encapsulate the various chips and components in a non-conductive polymer matrix. Obviously a liquid would work well for this process, as it would easily flow around the components, and then could be designed to cure into a polymeric matrix. Many of the assembly processes however are not well suited to the presence of liquid chemicals that would need to be dispensed and distributed to a tightly controlled thickness. A PSA film, therefore, has some significant advantages: ease of handling, guaranteed uniform thickness, and the potential to have the right viscoelastic properties to flow around the chips and circuit components to provide suitable encapsulation.

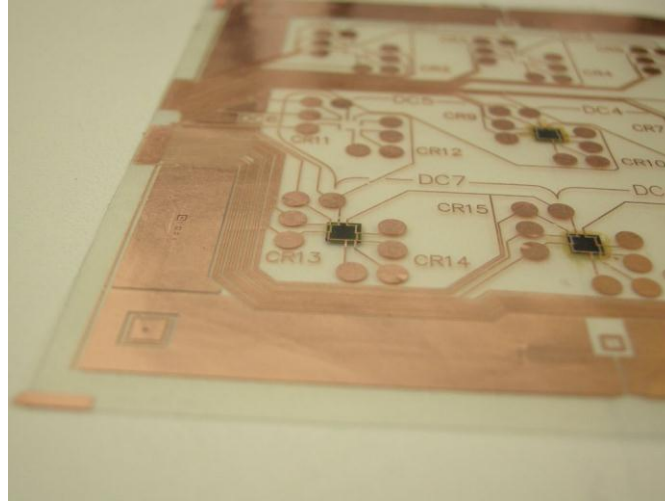


Figure 7. Flexible printed circuit.

The process we envisaged for the use of a PSA film is outlined in Figure 8. Conductive adhesives (non-PSA) are dispensed onto a copper foil (step 1), and the chips are attached (step 2). A film is then placed on top that is constructed of a PSA, of perhaps 50 micron thickness, on a film backing, perhaps a 50 micron or less PET (step 3). The lamination process allows the PSA to flow suitably around the chips. After that, lithographic etching and other processes remove some of the copper to expose areas for further electrical connections in the structuring step (step 4).

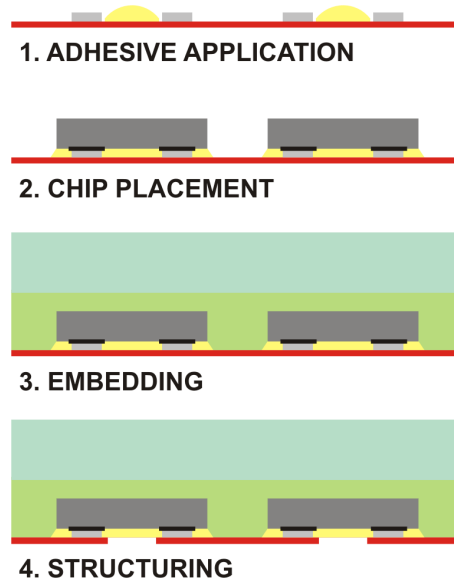


Figure 8. Chip embedding process.

Our work to date has shown that this process can be accomplished reasonably well with existing, commercial acrylic PSAs. The viscoelastic nature of many PSAs allows for suitable flow around the chips and other components. It is important to control the low levels of volatiles in the adhesive, so as not to cause outgassing or the formation of

bubbles in the adhesive under subsequent heat exposure and processing. While the products we have been using do not always give perfect laminations, we have been able to produce suitable demonstrators and working components, using commercially available PSAs. Figure 9 below shows one such demonstrator.

Development work continues in this area, focused largely on the lithography step at the end of the process. In some instances, this can result in areas of the PSA being exposed, when the copper foil is etched away. Having an exposed adhesive surface is not necessarily beneficial, and it may be useful to have an adhesive that behaved as a PSA during the initial lamination and then underwent a secondary curing step to become more like a thermoset material, with limited or no PSA properties after the etching was complete.

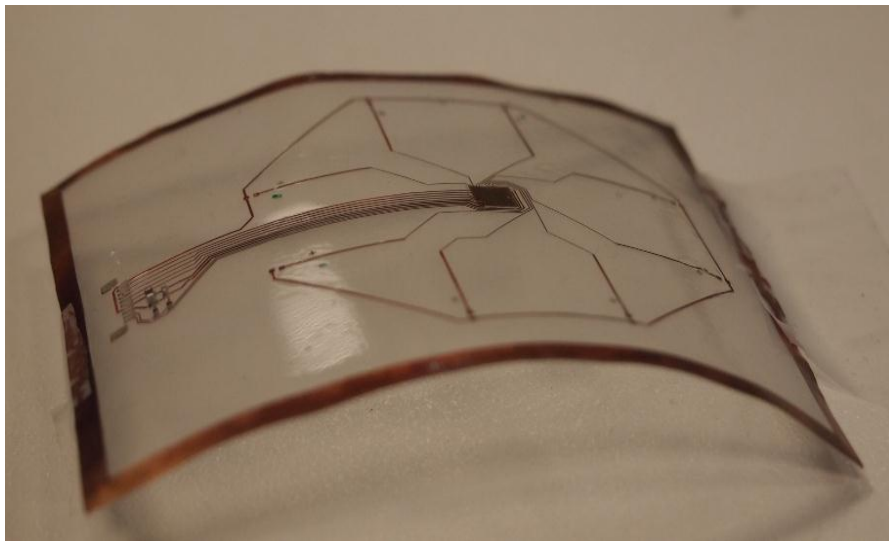


Figure 9. Flexible circuitry with PSA encapsulation.

Barrier adhesives

Arguably, one of the most important material properties needed to allow the large scale, commercially viable creation of flexible displays using printed OLEDs is the creation of barrier materials.

OLEDs are air and moisture sensitive, and exposure to either will cause degradation and eventually destruction of the OLED. This will result initially in black spots on the display, and over time will cause complete failure of the display. It is partly due to this sensitivity that most commercial OLED displays today are created on glass substrates, as the glass provides an excellent barrier.

If an OLED were to be produced on a flexible substrate, then a rigid glass barrier can no longer be used. Instead, flexible films and foils would form the outer layers of the display. Flexible OLEDs have been made in sheet form, in batch processes, where the

entire assembly is shrouded in a barrier material after manufacture. Whilst successful in stabilizing the OLED, this is not practical for a low cost roll-to-roll process. One alternative is to use existing adhesives to form the OLED laminate, and then encapsulate the edges of the structure, as shown in Figure 10.

Many of the existing encapsulants are not particularly flexible, and should this structure suffer any delaminations at the edges, then the entire OLED will fail, as moisture and air could begin to access the bond lines in the laminate.

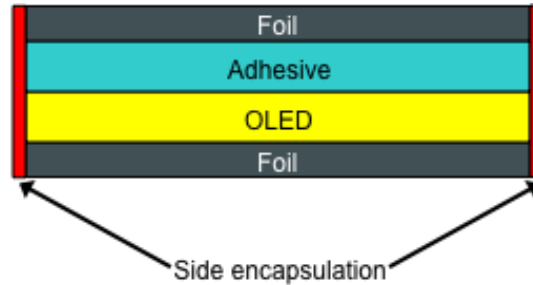


Figure 10. Side encapsulation of OLEDs.

Perhaps a better solution would be to create a structure where the adhesive itself had suitable barrier properties, and side encapsulation was no longer necessary, such as is shown in Figure 11.

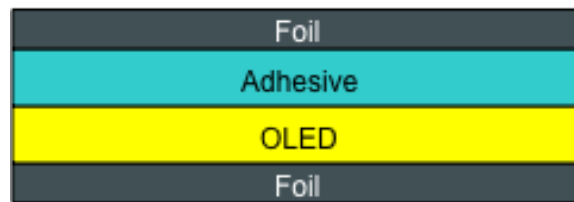


Figure 11. Barrier adhesive on OLED.

This is the direction we have taken in our work – to attempt to create an adhesive that can bond the layers of the OLED structure together, be chemically compatible with the OLED, and provide suitable barrier protection to prevent ingress of moisture from the side of the construction.

Acrylic PSAs are not suited to these applications, as their moisture permeation is much too high. Instead we turned to polyisobutylene adhesives. These offer both much better barrier properties than acrylics as well as being non-functional, and so less likely to cause chemical interactions with the OLEDs.

The low glass transition temperature of polyisobutylene gives it many inherently useful PSA properties. One disadvantage, however, to many commercially available PIBs is that their non-functional nature makes it difficult to build shear strength into the adhesive, which can limit their usefulness in many potential applications.

Our approach involved using commercially available PIB polymers, in solution, and formulating them with various functional materials and UV crosslinkers, to enable the production of a coating that would have the expected barrier properties of a PIB, coupled with the cohesion needed for the application. As with the polarizer adhesives discussed earlier, we have been coating from solution and then curing by UV radiation. This gives us both the coating quality we need, and the rapid cure, which is beneficial in the application.

The moisture uptake rate of the adhesives we designed was between 60 and 100 ppm when measured at 23°C and 50% relative humidity with a water diffusivity coefficient of approximately $0.4 \times 10^{-12} \text{ m}^2/\text{s}$ under the same conditions. For reference, values for the diffusivity coefficient of (meth)acrylates are considerably higher – approximately $13.3 \times 10^{-12} \text{ m}^2/\text{s}$ for PMMA, for example⁷.

To test the ability of our PSAs to provide barrier properties, we started by using rigid OLEDs. Ultimately, our goal was to create flexible systems, but the rigid systems would allow for barrier property evaluation separate from the potential complication of a flexible system.

Our test configuration used a plate with multiple test sites as shown in Figure 12. The layout on the plate consists of nine OLEDs, and allowed for testing of three configurations, with each one tested in triplicate. The configurations used were:

- Adhesive with a getter (top row)
- Adhesive without a getter (middle row) and
- No adhesive or getter (bottom row)
-

The last row acted as a control, to examine the decay of the OLED without any additional layers or barriers.

The getter is a moisture-absorbing matrix (typically calcium oxide) that absorbs, but does not release, moisture.

Samples prepared in this way were then subjected to various tests. Firstly, we ran JVL curves (current density – voltage – luminance) both before and after the application of the adhesive to the OLED. These data are shown in Figure 13, which indicates no meaningful change in performance of the OLED before or after the application of the adhesive. This leads us to conclude that the adhesive is chemically compatible with the OLED and that the efficiency of the OLED is unaffected by the presence of the adhesive.

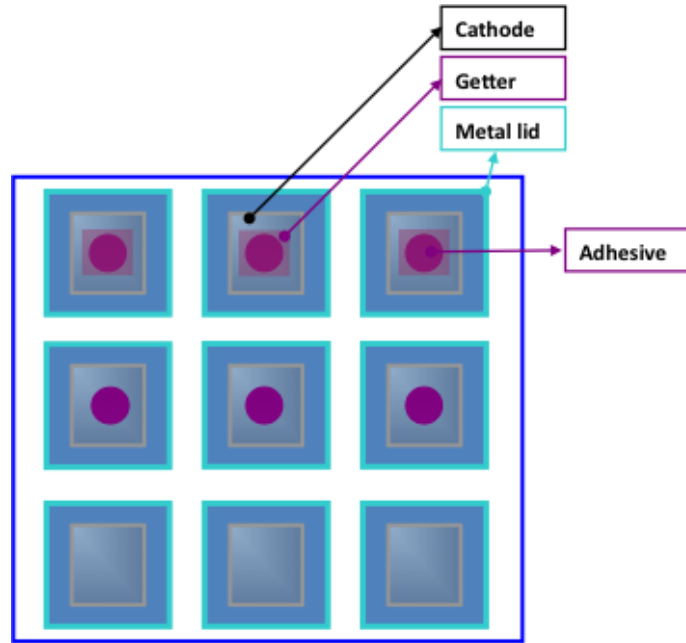


Figure 12. OLED test assembly.

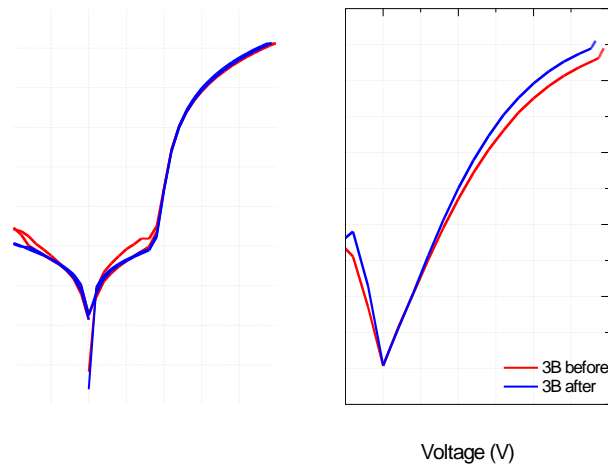


Figure 13. JVL curves.

Testing then continued by aging the assembled OLED plates at 60°C and 90% relative humidity and examining the samples regularly for the presence of black spots – an indication of OLED degradation.

Whilst the examination and counting of black spots over time was done using a microscope connected to a digital analyzer and a computer program, the most striking results are from a visual inspection of the samples. As Figure 14 shows, the adhesive provides significant protection the OLED against decay. The bottom row of images

shows substantial OLED degradation after 12 days (288 hours), whilst the performance is markedly better with the adhesive in place (and as expected, even better when a getter is used).

We do see some degradation of the OLED after 12 days, and obviously this is not suitable performance for a commercially viable OLED, where the display would be expected to last for multiple years. It is, however, an impressive start, and on a system with no edge encapsulation, meaning that the adhesive alone was providing protection on the side of the assembly.

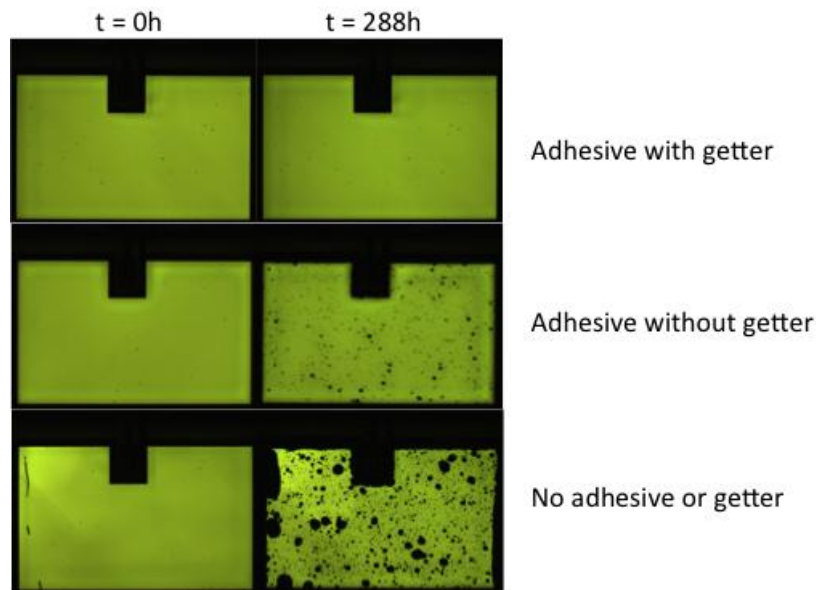


Figure 14. OLED stability tests.

There is obviously much work still to be done, as we improve our barrier performance and extend our testing to flexible systems, but we are encouraged by these early results and remain committed to understanding the adhesive needs of these applications and developing products that help to realize the commercialization of the technology. Holst has already produced some demonstration OLED samples, although with very short lifetimes. By the end of 2012, we intend to have a working demonstrator using our adhesives that will have a useful working lifetime.

Conclusions

This paper has described some of our initial results in our efforts to understand technology areas that move beyond many of our traditional applications. We have a lot of work still to do, but we believe that extending our PSA capabilities into areas such as the display applications we described here offers exciting possibilities for the future and will help to grow and improve the applications and markets available to PSA coaters and users.

Acknowledgements

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