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ORGANIC PHOTONICS

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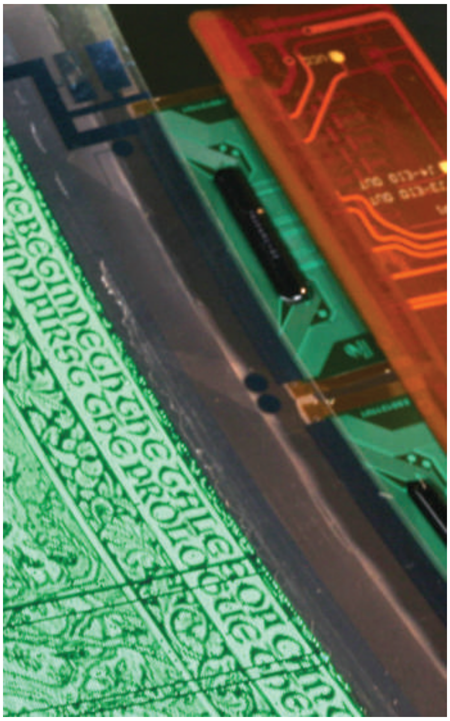

COVER IMAGE

Active-matrix OLEDs provide efficient and flexible display solutions. (Image credit: Flexible Display Center)

The age of organics

After a long and arduous development process, lighting products and large displays based on organic light-emitting diode (OLED) technology are finally coming to market. In our product highlights on page 457, the Korean electronics giant Samsung reveals that it has now developed 14.1- and 31-inch OLED television panels that are ready for commercial production. Not only are these displays very thin but they also boast impressive brightness and colour fidelity, suggesting that they could become a feasible alternative to traditional liquid crystal displays.

It is new material technology, such as the doping schemes from Novaled (see page 444), that has helped improve the performance of small-molecule OLEDs in recent years, and polymer OLED technology is not far behind according to Cambridge Display Technology (see page 453). However, challenges in both markets remain. For example, Marc Baldo, an expert in organic photonics from Massachusetts Institute of Technology, USA, believes the efficiency of blue OLEDs could still be improved (see page 458). He also points out that one of the major challenges facing OLED displays and the lighting industry is not actually the OLED materials themselves, where huge improvements have been made to efficiencies and lifetimes; surprisingly, the problem lies with the stability of the silicon backplane electronics. Twenty years ago, when researchers worldwide were struggling to achieve reasonable lifetimes and efficiencies from OLED materials, few would have predicted that silicon technology would be the bottleneck to commercialization.

Another industry that has benefited from the advances made in OLED technology is the organic photovoltaics sector. Major improvements have been made, with efficiencies above 6% reported for the first time (see page 447). However, as with the OLED industry, and indeed any fledgling market, many challenges remain: examples include developing reliable manufacturing processes and further improving the lifetimes of these low-cost, flexible cells. However, if the progress of the last 10 years is anything to go by, these challenges may be short-lived.

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DuPont's OLED material hits million-hour lifetime

DuPont Displays has developed a solution-based green OLED material that has a lifetime of over 1,000,000 h. The company claims its third-generation OLED materials can meet or exceed the performance of today's vapour-deposited materials, and are paving the way for low-cost solution-processed OLED displays.

"Printing OLEDs significantly lowers the cost of manufacturing displays, and with Gen 3 materials technology, display manufacturers can see the material lifetimes and performance required for commercialization," said William Feehery, global business director for DuPont OLED Displays. "With lifetimes five times better than just two years ago, these new materials will allow solution OLEDs to be used in mobile displays, and also to begin to penetrate the television and general lighting markets at a lower cost than today's evaporated OLED technology."

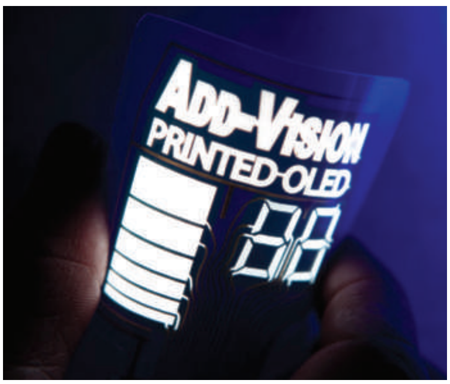
DUPONT DISPLAYS

The new Gen 3 green material developed by DuPont has a lifetime of 1,000,000 h — a milestone in performance that equates to over 100 years of constant use. This green material demonstrates a light emission efficiency of 25 cd A^{-1} and excellent colour coordinates (0.26, 0.65).

Historically, improving the performance of blue light-emitting materials has been the biggest challenge; however, DuPont Gen 3 blue materials are also achieving good results. One of the Gen 3 blue materials has colour coordinates of (0.14, 0.12), a light generation efficiency of 6.0 cd A^{-1} and a lifetime of 38,000 h with an initial brightness of 1000 cd m^{-2}. Another blue material has been developed with exceptionally deep blue colour coordinates of (0.14, 0.08), an efficiency of 3.9 cd A^{-1} and a lifetime of almost 7,000 h. Owing to its deep blue colour, the lifetime of this material at the luminance required for a 200 cd m^{-2} display is calculated to be approximately 41,000 h.

Furthermore, a high-performing red solution OLED material developed by DuPont has a lifetime of 62,000 h, current efficiency of 13 cd A^{-1} and colour coordinates of (0.68, 0.32).

Bayer and Add-Vision sign polymer OLED agreement



ADD-VISION INC

Polymer OLED (P-OLED) developer Add-Vision Inc has signed a technology and patent license agreement with materials supplier Bayer MaterialScience. The agreement grants Bayer MaterialScience and its affiliates certain rights to manufacture and sell flexible P-OLED displays using Add-Vision's technology and intellectual property portfolio. Financial details, however, have not been disclosed. Karsten Dierksen, vice president and head of the Functional Films Polymer Electronics Unit at Bayer MaterialScience, said: "We are pleased to add this promising technology to our Functional Films portfolio. It's a great fit as it perfectly complements our electroluminescence technology." Matt Wilkinson, Add-Vision's CEO added: "This important agreement with Bayer MaterialScience is the starting point for a strong relationship. Our partner's commitment to film-based technologies and products, combined with the print-based manufacturing expertise of its Functional Films Unit, makes this an ideal collaboration."

Future applications include active packaging and labels, electronic toys and games, as well as point-of-sale signage.

Kodak bags $1.7 million lighting project

A contract worth $1.7 million has been awarded to Eastman Kodak by the United States Department of Energy (DOE) for a two-year contract to develop OLED lighting panels. As a pioneer in OLED technology, the Rochester-based company has recently exceeded the project efficiency target set by the DOE of 50 lm W^{-1}. Kodak's latest OLED delivers an efficiency of 56 lm W^{-1}, a lifetime of 10,000 h, a colour rendering index of 83.6 and a colour temperature of 4,000 K. Kodak says that one of its main objectives now is to reduce manufacturing costs. The firm is developing all-fluorescent and hybrid systems with the aim of achieving longer lifetimes than all-phosphorescent emitter systems. Thanks to its proprietary 'vapour injection source technology', Kodak says that it can reduce manufacturing costs by increasing material utilization and reducing fabrication time. It expects to see a cost reduction of up to 50% and, ultimately, perhaps even 75% in the future.

Polytos project to aid organic electronics

A cluster comprising 27 companies, universities and research institutes in Germany has recently launched the 'printed organic switches and chips' (Polytos) project. Sponsored by the German Federal Ministry of Education and Research and headed by Merck KGaA, the project aims to develop new materials, concepts, components, manufacturing processes and software for printed organic circuits. The consortium says that it is trying to achieve low-cost printed organic circuits that can function as 'smart labels', capable of recording data (such as temperature, humidity or light exposure) on an industrial scale in the future. The project is expected to last three years and cost around €13.8 million, with €7.2 million from the German Federal Ministry and €6.6 million from industry partners.

With the knowledge and involvement of experts ranging from researchers to those in industry, the consortium looks set to help expand Germany's international leadership role in the development of organic electronics.

C_{70} boosts organic photovoltaics

[1]*Appl. Phys. Lett.* **94,** 223307 (2009)
[2]*Sol. Energy Mater. Sol. Cells* **93,** 1149–1153 (2009)

Organic solar cells (OSCs) are a promising solution to cost-effective solar energy generation, owing to their potential for lower production and material costs when compared with inorganic approaches. However, the efficiency of OSCs must improve if they are to become a practical alternative. Fullerene C_{60} is commonly used as the charge transporter and acceptor in small-molecule OSCs, but its absorbance in the visible spectrum is believed to limit cell efficiency. Two teams have now shown that fullerene C_{70} could be an attractive alternative. Steffen Pfuetzner and his collaborators at the Technische Universität Dresden, Germany, showed that the larger fullerene exhibits higher absorption, particularly in the visible range of 500–700 nm (ref. 1). The associated increased photocurrents gave rise to an overall efficiency of 2.87%; this represents an increase of 26% over C_{60}-based devices (efficiency of 2.27%) with similar fill factors and open-circuit voltages. Simultaneously, Jun Sakai at Panasonic Electric Works in Osaka, Japan and his colleagues at the National Institute of Advanced Industrial Science and Technology in Ibaraki, Japan, have shown that the performance characteristics of C_{70} can be enhanced by thermal annealing[2]. The annealing process yields increased molecule packing and 40%-reduced series resistance (and hence superior current-voltage characteristics) in solar cells.

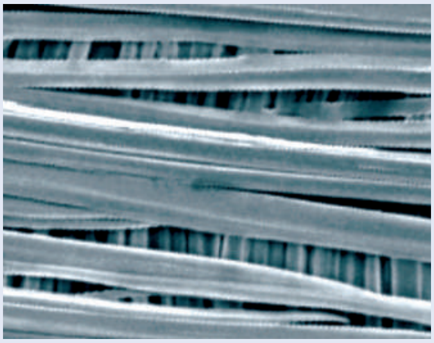
A polyfluorene blue semiconductor laser

Appl. Phys. Lett. **94,** 253304 (2009)

Blue-emitting polyfluorene (PFO) is known to be a useful laser gain medium, owing to its low threshold and tunability. Unfortunately, PFO, in common with most organic blue emitters, absorbs in the ultraviolet spectrum where there is a lack of convenient pump sources. Researchers at the University of St Andrews have overcome this problem by making a blue organic semiconductor laser that is instead pumped by red light. Using a red pump wavelength of 640 nm, the team excited two-photon fluorescence in the PFO to yield blue emission in the range of 442–458 nm. The team made the blue-tunable solid-state PFO laser device by spin-coating a PFO solution onto silica substrates with corrugated surfaces. The periodic surface relief provided the required distributed feedback for the laser cavity. A lasing threshold of 42 mJ cm^{-2} at an absorbed energy density of 1.3 mJ cm^{-2} was demonstrated in the two-photon scheme. The results highlight the potential for using nonlinear pumping schemes for the development of compact polymer lasers.

Nanowire films in order

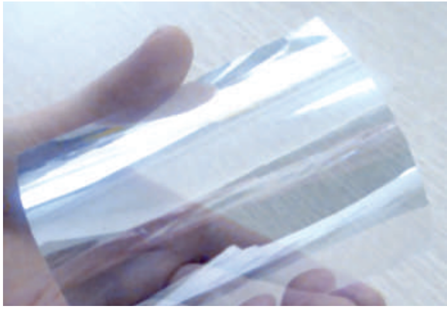
Adv. Mater.
doi:10.1002/adma.200802793 (2009)



One-dimensional organic nanomaterials are potentially attractive for building miniature optoelectronic devices. Before they can be put to use, however, cost-effective fabrication and assembly processes are essential. Now, Chengyi Zhang and colleagues from China have demonstrated a simple one-step process for growing single-crystal organic nanowires and assembling them into aligned films. The trick is to perform the fabrication at a liquid–liquid interface between dichloromethane (DCM) and water. By evaporating the liquid solvent, DCM, organic nanowires form and then assemble into ordered films, owing to a compression force that results from the shrinking DCM–water interface. Zhang and colleagues say that the resulting films can be easily transferred to a substrate or stacked to form a multilayer film for device applications. As a proof of concept, the researchers prepared a film of nanowires doped with squaraine dye and used it to create a photodetector and a field-effect transistor. Excellent photoresponse and stability were obtained, demonstrating the versatility and scalability of this fabrication technique for forming large-scale, compact films of organic nanostructures at low cost.

Flexible OLEDs offer polarized output

Opt. Express **17,** 10136–10143 (2009)



By replacing the usual glass substrate with a polarizing polymer material of large optical birefringence, researchers in South Korea have succeeded in making a green organic light-emitting diode (OLED) with a bright and strongly polarized output. Byoungchoo Park and colleagues from Kwangwoon University report that their OLED has a luminous emission of 4500 cd m^{-2} and a polarization ratio as high as 25. Peak efficiency of operation was in excess of 6 cd A^{-1} and 2 lm W^{-1}. The transparent polymer film substrate was 90 μm thick and functioned as a strong polarizer in reflection. It is a commercially available product manufactured by the company 3M. The researchers conclude that the results allow the realization of a new class of thin and flexible polarized OLEDs for applications that require a polarized light source, such as backlights for liquid crystal or three-dimensional displays.

OLEDs overtake fluorescent tubes

Nature **459,** 234–238 (2009)

White OLEDs with a power efficiency exceeding that of fluorescent tubes suggests that OLEDs may have an important role in reducing the energy demands for lighting. Writing in *Nature*, Sebastian Reineke and co-workers from the Institut für Angewandte Photophysik, Germany, reveal that they have fabricated OLEDs with a record efficiency of 90 lm W^{-1} when operating at 1000 cd m^{-2}. In comparison, typical fluorescent tubes have an efficiency of 60–70 lm W^{-1}. Furthermore, the researchers believe that it should be possible to increase the efficiency of their OLEDs to 124 lm W^{-1} if the light outcoupling from the devices can be further improved. The key to the performance of the phosphorescent OLED is the optimal positioning of a blue phosphor layer within the design and the careful choice of host material that combats unwanted non-radiative relaxation. The big challenge now is to reduce the cost of manufacturing through the use of low-cost electrode materials, thin-film encapsulation and roll-to-roll manufacturing.

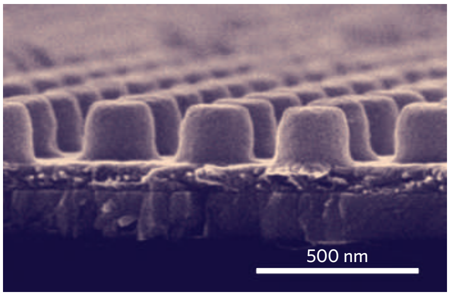
Light-emitting sensors

Adv. Mater. doi:10.1002/adma.200802237 (2009)

In addition to emitting light, OLEDs can also be used to construct small and convenient gas sensors. This is the finding reported by Stefan Sax and co-workers from Austria who have demonstrated that organic semiconductors can help the sensing of an analyte. The team constructed a device with a similar structure to a polymer-based OLED, except that an analyte-sensitive dye molecule was added to the OLED's electro-active layer. The researchers observed a decrease in emission intensity when the device was exposed to oxygen. They confirmed that the luminescence decrease does not relate to the degradation of the device during operation but is instead attributed to the quenching of excited dye molecules in the electro-active region as a result of oxygen diffusion. Because a wide variety of dye molecules can be used for this set-up, the researchers anticipate its use beyond oxygen detection.

Photonic crystals boost efficiency

Nano Lett. **9,** 2742–2746 (2009)



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Photonic crystals were investigated with the aim of improving organic solar cell efficiency. New benefits have been seen experimentally by a group of researchers from the University of North Carolina in Chapel Hill, USA. Doo-Hyun Ko and colleagues imprinted a two-dimensional photonic crystal structure into the photoactive bulk heterojunction layer of organic solar cells. As a proof of concept, a blend of thermally deprotectable polythiophene derivative (TDPTD) poly(3-(2-methyl-2-hexylcarboxylate) thiophene-co-thiophene) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) were used as the bulk-heterojunction layer. The photonic crystal structure was composed of highly ordered arrays of columnar features with a diameter of 110 nm and a periodicity of 400 nm. It was fabricated in the layer using pattern replication in non-wetting templates (PRINT) in a single process over areas of ~4 cm^2. The team obtained a threefold absorption enhancement near the band edge of the bulk-heterojunction layer and an efficiency improvement of ~70%. They attributed these results to improved absorption and electrical enhancements due to the photonic geometry. Owing to the multiple resonances in the photonic crystal structures, the approach allows selection of desired regions of the solar spectrum for absorption enhancement.

Surface plasmon enhancement

Opt. Express **17,** 11495–11504 (2009)

Surface-plasmon-enhanced absorbance and emission enables more efficient energy transfer in organic light-emitting devices, according to Ki Youl Yang and co-workers from South Korea. The researchers studied the properties of an Alq_3:DCM-based fluorescent OLED, fabricated by vacuum thermal evaporation. To achieve localized surface plasmons, Yang *et al.* deposited a silver nano-cluster thermally close to the cathode with a 1-nm-thick LiF spacer. They observed 3.5-fold enhanced acceptor emission intensity in photoluminescence, compared with device samples without the silver nanocluster. The enhancement was attributed to the strong donor decay channel and increased donor–acceptor dipolar interaction, induced by surface plasmons. The team also pointed out that the surface plasmon excitation wavelength should be set close to both the donor emission and acceptor absorbance peaks to enhance the energy transfer in the donor–acceptor system. In particular, they found that the donor decay rates increased more as the wavelength approached the donor emission energy, and the donor–acceptor interaction was enhanced as the localized surface plasmon energy approached the acceptor absorbance centre. This scheme helps increase the emission efficiency of OLEDs and provides a useful mechanism for advancing the performance of optoelectronic devices and biosensors.

Ultrafast all-optical modulation

Appl. Phys. Lett. **94,** 253301 (2009)

Polymer films containing copper porphyrin tape are useful for performing ultrafast all-optical light modulation in the near-infrared region. That is the recent finding of Ryuji Matsumoto and colleagues from Kyushu University and Kyoto University. Unlike previous modulation methods that rely on changing the real part (Δn) of a material's complex refractive index using an electric field, heating or photochromism, the Japanese team instead changed the imaginary part (Δk) of the refractive index using light. They achieved this by taking a glass prism and spin coating one side of it with a 600-nm-thick low refractive polymer film (Cytop) followed by a 400-nm-thick polymer film containing copper porphyrin tape tetramer (CuT4). The polymer films are optically pumped and interact with a polarized signal beam that is coupled into and out of the prism via its uncovered faces. The modulation effect is due to photoinduced transient absorption in the polymer film containing CuT4, which, thanks to the Kramers–Kronig relation, relates to a change in Δn. By having an appropriate combination of the polarizer and the analyser in the set-up, the researchers reported that optical modulation of output intensity could be achieved. For a wavelength of 1100 nm, the response time of the transient absorption was measured to be less than 1.3 ps.

Magnetic field improves OLED intensity

Jpn. J. Appl. Phys. **48,** 061502 (2009)

Although the output emission intensity of an OLED often decreases gradually over time, researchers in Japan have discovered that in fact the opposite occurs — intensity actually increases if a magnetic field is applied to the device. Hiroshi Okimi and co-workers from RIKEN, Tokyo Institute of Technology and Sumitomo Chemical, applied an external magnetic field of up to 800 mT to OLEDs made from a luminescent layer based on poly(1,4-phenylenevinylene) (PPV). They observed that at a field strength of ≥200 mT, the emission intensity was 1.4–1.5 times higher than with no applied field. They also observed that the emission intensity increases over time, reaching a maximum after approximately 20 h. The increase in emission intensity is attributed to the promotion of long-range charge recombination, induced by a structural change in the OLED material.

Small company, big plans

German company Novaled has built a business around a doping technology that increases the efficiency of organic LEDs. Its materials have broken many efficiency records and are being used in a wide range of applications, reports **Nadya Anscombe**.

When a group of researchers from the Technical University (TU) of Dresden, Germany, established Novaled in 2001, the market for organic light emitting diodes (OLEDs) looked very different from today. There was a clear distinction between companies developing polymer OLEDs and those developing small-molecule OLEDs; both camps were competing to be the first to commercialize their technologies and it was unclear which technology was most likely to succeed.

Jan Blochwitz-Nimoth and his co-founders were interested in developing small-molecule OLEDs and believed they had found a way of making them more efficient — a commercially viable doping technology (Box 1). This caught the attention of the venture-capitalist community, which has invested a total of €28.5 million in the company so far — one of the largest venture-capital investments ever made in a German technology company.

Although many other OLED developers have been bought and sold repeatedly, Novaled has retained its original founders and seen its revenues and workforce steadily increase. "In 2008, the company generated revenues of US$10 million," says Blochwitz-Nimoth. "We want to further develop the OLED lighting and display market and aim to break even next year."

The company has recently set up a facility in Japan and can no longer be described as simply 'a developer of small-molecule OLEDs'. The techniques for printing polymers and vacuum depositing small-molecule compounds have converged, and so Novaled is now working together with companies such as Cambridge Display Technology/Sumation and Universal Display Corporation to develop the most appropriate materials and manufacturing processes for each application.

"A small company such as Novaled can't do everything," admits Blochwitz-Nimoth. "So we are working with several industry partners to develop applications and commercialize our technology."

Novaled has chosen a 'fabless' business model; it does not manufacture products, but instead generates revenues primarily from royalties by selling technology access and materials. Its proprietary materials are available in large quantities from its industrial partner BASF (formerly Ciba). "This ensures the highest quality materials and on-demand availability for our customers," says Blochwitz-Nimoth.

Jan Blochwitz-Nimoth, one of the co-founders of Novaled, says that the firm's technology not only allows the creation of bright displays and lighting, but also efficient solar cells and flexible electronics.

The company is a key player in projects such as OLED100 (www.oled100.eu), a three-year pan-European research project that has received €12.5 million from the European Community's Seventh Framework Programme. It aims to develop an OLED technology that can achieve a 100 lm W^{-1} power efficiency, more than 100,000 'lifetime hours', a unit area of 100 cm × 100 cm and costs less than €100 per square metre.

Over the years, the Novaled team has broken many records for power efficiency and claims to still hold one for green OLEDs (163 lm W^{-1} at 1000 cd m^{-2} initial brightness).

"Researchers at the TU Dresden using our technology recently achieved a power efficiency of 90 lm W^{-1} at a brightness of 1000 cd m^{-2} for a flat lighting device, and even 124 lm W^{-1} when using a 3D light extraction system," says Blochwitz-Nimoth. "This even beats the efficiency of a fluorescent tube. At a very high brightness of 5000 cd m^{-2} a power efficiency of 74 lm W^{-1} is obtained, making high-intensity illumination viable too." This latest performance was reported recently (*Nature* **459,** 234–238; 2009).

Results like this mean that Novaled's PIN OLED technology (Box 1) enables the production of displays with lower power consumption, longer lifetimes and better temperature stability than other OLED technology. "Power efficiency is crucial for portable devices, but also for large screens because of heat dissipation," says Blochwitz-Nimoth. "In addition, we offer high flexibility for our customers and satisfy their need for inverted or non-inverted and top-or-bottom emission structures. Thus, our technology is suitable for all kinds of substrates. For example, our inverted top emission structure perfectly matches with amorphous silicon active-matrix backplanes, required for video applications and television."

However, he admits there is still work to be done. "Although our track record of record-breaking efficiencies clearly shows that OLEDs are suitable for mainstream lighting applications, we do realize these are lab results. Further development is needed; for example, to achieve commercially acceptable product stability before OLED lighting products come to market." The company is currently offering prototype lighting products and displays.

The OLED displays market is slightly more mature than the OLED lighting market; two important players in the displays market — Sony and Samsung — have launched OLED televisions in recent months. Novaled has been unable to comment on whether these products use its materials.

The displays market has, for many years, been the focus of attention for OLED development. Now that many of the OLED lifetime and efficiency targets have been achieved for displays, OLED companies can focus on lighting applications.

Many technical challenges, however, are common to both applications. For example, there are issues with the yield and reliability of the silicon backplane. In contrast to LCDs, OLEDs are an emissive technology, and this makes them sensitive to defects and power surges in the silicon backplane, causing problems in the display and lighting industries.

In response to this challenge, Novaled has developed defect-tolerant OLEDs. They maintain the appearance of a homogeneously-lit surface — even in the case of electrical short-circuits — owing to the use of a proprietary electrode design. One OLED element consists of two comb-shaped interlocked electrodes, giving a uniform light emission. If one of the stripes short-circuits, the resistance of the stripe will limit the current flow and prevent a further rise in current and temperature, which would otherwise destroy the OLED.

"OLED devices based on the new Novaled structure are well-suited for large-area lighting or other lighting applications that require a long maintenance cycle," said Blochwitz-Nimoth.

Novaled is also developing its technology for other applications, such as photovoltaics and organic electronics, and thin-film transistors (TFTs).

For example, Novaled's sister company Heliatek (also a spin-off from TU Dresden) is making significant progress in solar-cell development using Novaled's doping technology. It aims to achieve efficiencies of 8–10% over the next three years by producing solar cells that cover the entire solar spectrum. Using vacuum technology, these solar cells can be produced on a roll-to-roll basis, enabling manufacture of cheap, flexible cells.

"Our technology enables organic solar cells with the highest efficiencies, just as it does with OLEDs," says Blochwitz-Nimoth. "It allows easy integration of tandem architecture (stacking of two solar cells on top of each other) with excellent performance."

So far, 5% efficiency has been reported using Heliatek's tandem architecture (based on Novaled p- and n-dopants, combining absorber materials from Heliatek and BASF). The devices have a high open-circuit voltage (1.9 V), a high fill factor (60%), excellent thermal stability and a lifetime of up to 16,000 h (exposed to 100 mW cm^{-2} white light).

In organic electronics — transistors in particular — Novaled's materials are showing promising results. In organic TFTs, the carrier injection from the drain and source into the organic material has a major influence on device performance. The success of classical silicon complementary metal-oxide-semiconductor (CMOS) technology is based on the combining of p-type and n-type silicon transistors, and similar organic complementary circuits can be manufactured by using Novaled's controlled p- and n-doping technology. Both p- and n-type field-effect transistors can be made side-by-side based on a single semiconductor layer, by controlled p- and n-doping of the contact regions. This enables engineers to reduce contact resistance at the source/drain, use less expensive contact materials (replacing gold), define conduction type for a wide range of materials, increase mobility and control threshold voltage.

The successful development of organic TFTs will lead to flexible electronics that will revolutionize many different industries. As well as electronic structures, another essential step towards this goal is thin-film encapsulation to replace the glass substrates that are currently used. In this area, Novaled has cooperated with Sunic System and Vitex to develop turn-key solutions and materials for thin-film encapsulation technologies, targeting ultra-thin OLED devices.

With major companies unveiling OLED TVs and lighting product prototypes at every trade show, it is clear that the future is bright for OLED technology and Novaled. ❒

The Victory Lamp is a prototype lighting product that aims to demonstrate how Novaled's OLED technology can be used. The lamp features 5 cm × 5 cm OLEDs and gives off a warm white light.

Box 1 | Doping expertise

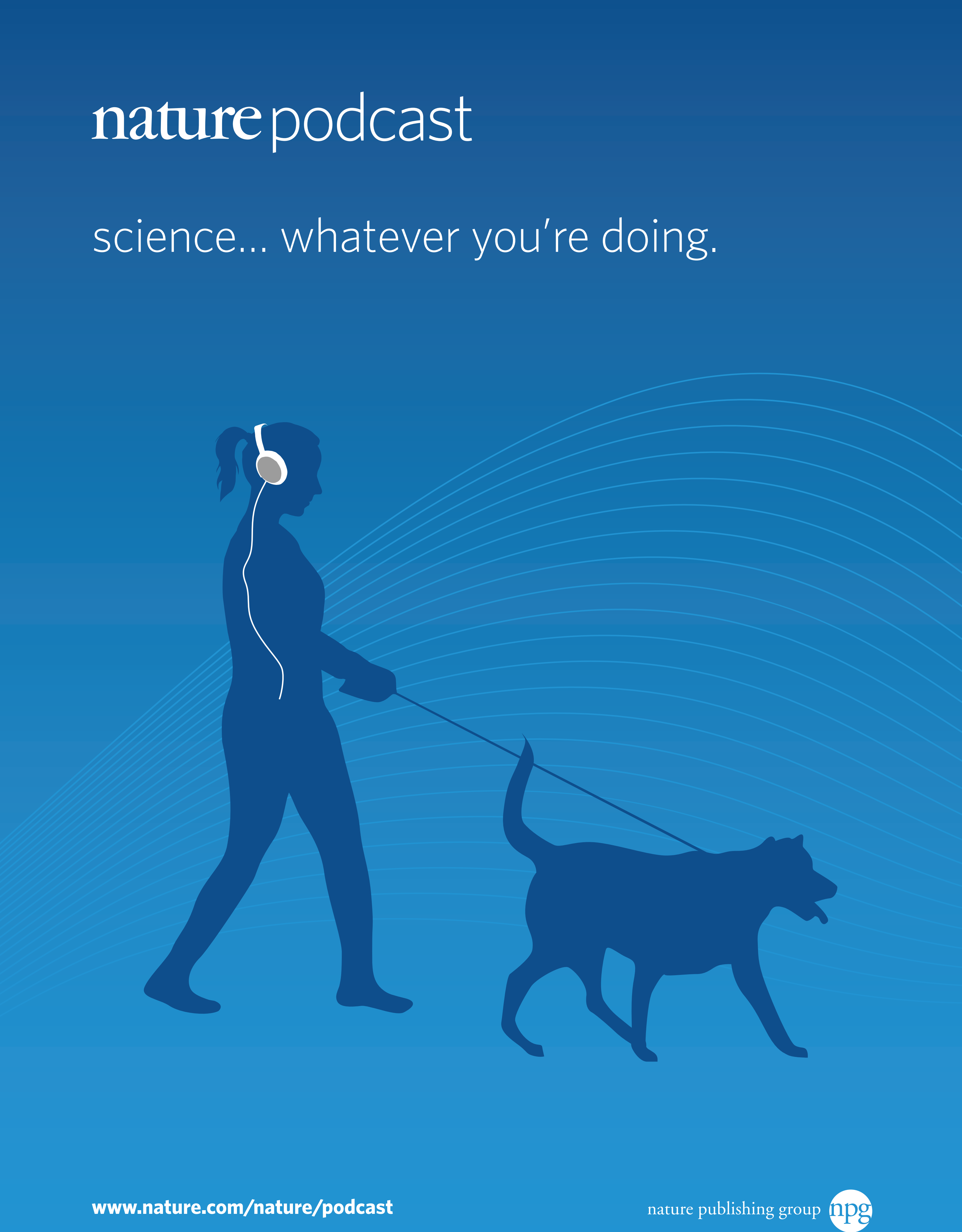
The power efficiency of OLEDs depends primarily on the operating voltage and the current efficiency. Novaled's products require only 50% of the power required by other OLED devices currently being developed.

Novaled's key technology is the improvement of charge-carrier transport. This is achieved by doping the charge-carrier transport layers with proprietary dopants to increase the conductivity and optimize the structure of the OLED, achieving a low operating voltage and high efficiencies at high brightness levels. The company claims that this doping approach can be easily implemented by co-deposition in today's OLED technologies. The doping technology also enables thicker charge-carrier transport layers, which are less susceptible to leakage currents or voltage breakdown, and thus enhance the reliability of OLED devices.

Novaled's technology is based on a p-i-n type OLED structure with extremely low operating voltage and high efficiency. This p-i-n OLED (or PIN OLED) structure consists of a p-doped hole transport layer (p-HTL), an intrinsic electron-blocking layer (EBL), an emission layer (EML), a hole-blocking layer (HBL) and an n-doped electron transport layer (n-ETL). The p- and n-doping results in high conductivity (in the range of 10^{-5} S cm^{-1}) and a Fermi-level shift, leading to high current-injection from both electrodes into the organic layers.

Novaled's doping technique enables flexibility in the diode thickness, allowing the out-coupling of light to be optimised by the diode thickness without influencing the number of photons generated.

Approximately 80% of the excited light is either trapped inside the OLED device or leaves in an undesirable direction. Therefore cost-efficient solutions for out-coupling of light — for example surface modification of the substrate — contain enormous potential for improving the efficiency of current flow. Another advantage of the technology is that it allows for thicker charge-carrier transport layers and wider process windows, further increasing yields.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ORGANIC PHOTOVOLTAICS

Polymer power

Vishal Shrotriya

With efficiencies continually improving, polymer solar-cell technology is now leaving the lab and entering the marketplace. Many challenges remain, however, including the development of reliable manufacturing processes and improvement of the lifetimes of these low-cost, flexible cells.

Solar cells based on polymers are considered a promising alternative to their inorganic counterparts for the generation of affordable, clean, renewable energy. Advantages of polymer solar cells (PSCs) include low-cost fabrication of large-area devices; low specific-weight; mechanical flexibility; easy tunability of chemical properties of the organic materials; attractive colours; better performance in low and indirect light; and the potential for transparent solar-cells.

As a result of these properties, PSCs can be used in applications that were previously unfeasible, such as incorporation into textiles or their use as rollable charging pads for portable electronics (Fig. 1). This article presents some of the recent advances in PSCs, including new polymers developed for higher efficiency, translucent solar cells and their applications, the commercialization potential of polymer PSCs, technical challenges that need to be overcome, and the future outlook for the technology.

Figure 1 | Prototypes developed at Solarmer energy, where polymer solar cells are integrated with consumer products. **a**, A messenger bag. **b**, A GPS charging pad. The cells will generate power under light, extending the battery life of portable electronic devices and allow users longer periods between charging.

There has been much interest from academia and industry in pushing to increase the efficiency of PSCs to a commercially viable level, primarily through new polymer development. Owing to these efforts over the past decade or so, record cell power-conversion efficiencies in the lab have reached more than 6%. For example, Plextronics, based in Pittsburgh, Pennsylvania, reported cells with 5.98% efficiency, as certified by the National Renewable Energy Laboratory (NREL). Recently, Konarka Technologies, based in Lowell, Massachusetts, reported an NREL-certified efficiency of 6.4% for 0.8 cm^2 cells.

Solarmer Energy, based in El Monte, California, recently reported an NREL-certified world-record PSC efficiency of 6.77% and a polymer solar panel efficiency of 3.9%, certified by Newport Corporation, California. All of these developments are exciting because they push PSCs towards commercial viability. However, despite this progress, further improvements in efficiency still need to be made before PSCs can be put into practical applications at an affordable price, which has long been the goal of this technology.

There is substantial debate over what the required threshold efficiency is for a successful commercial product. A growing number of researchers agree that an efficiency of 10% in the lab and 5% in commercial panels would make PSCs a competitive solar technology

One of the biggest advantages of PSCs is how easily they can be fabricated. A typical bulk-heterojunction polymer solar-cell structure is shown in Fig. 2a. In such a cell, a polymer active-layer — consisting of a bulk-heterojunction of donor and acceptor molecules — is sandwiched between two electrodes. The donor and acceptor molecules form an interpenetrating three-dimensional (3D) bicontinuous network. The bottom electrode is a transparent conducting oxide (TCO) with a high work-function, such as indium tin oxide, which allows the light to pass through and acts as the anode. The top contact is a metal with a low work-function such as calcium or aluminium, which acts as the cathode. The donor polymer is the active component in the cell, and absorbs light, generates excitons and contributes to hole transport. The acceptor molecule, usually a derivative of C60 such as [6,6]-phenyl-C61-butyric acid methyl ester (PCBM), helps in exciton dissociation where the hole remains on the donor molecule and the electron is transferred to the acceptor. After dissociation, the charges are transported in their respective phases to be collected at the electrodes.

Several high-efficiency polymers have now been developed by Solarmer Energy, in partnership with Luping Yu at the University of Chicago and Yang Yang at the University of California, Los Angeles (UCLA). The monomers of these polymers were synthesized and purified using standard synthetic methods, and with consistent quality. All of the polymers can be synthesized with a high yield of more than 60% by using Stille or Suzuki coupling reactions. All the synthesis procedures have been verified as being suitable for scaling up production from the gram to kilogram levels, and PSCs based on these polymers have consistently achieved more than 6% efficiency in the lab.

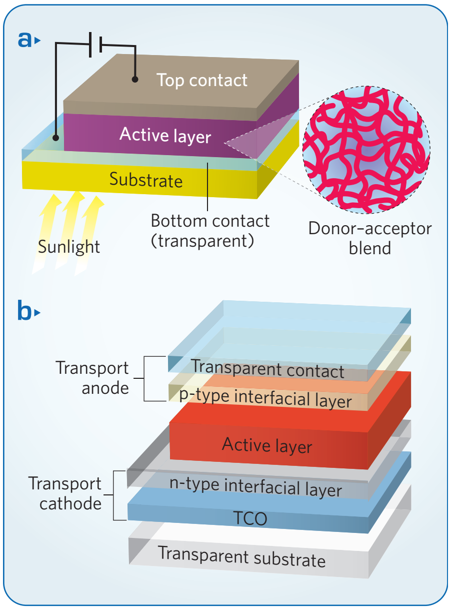

Figure 2 | Typical solar cells. **a**, Polymer bulk-heterojunction. The bottom transparent substrate can be glass or flexible plastic, the bottom contact is a transparent conducting oxide (TCO) with a high work function, the active layer is a mixture of donor and acceptor molecules, and the top electrode is a metal with a low work function, such as aluminium. **b**, Translucent polymer. The substrate is either glass or flexible plastic and the transparent cathode (bottom electrode) is made from a TCO coated with n-type interfacial film. The active layer is very thin donor–acceptor bulk-heterojunction film. For the anode (top electrode) a thin p-type interfacial layer is used and capped with very thin transparent conductive film, such as gold or a TCO.

Material research

One of the major challenges in achieving higher efficiency in PSCs is the lack of suitable absorber polymers with the required properties. New polymer development efforts have therefore been focused on improving the following properties:

Absorption. The large optical bandgap of polymers results in a mismatch between their absorption spectra and the solar irradiance spectrum. To increase efficiency, a polymer's absorption edge has to be increased to around 1,100 nm, to enable absorption in the infrared and near-infrared regions of the solar spectrum.

Exciton separation. Energy loss during the exciton separation process between donor and acceptor due to a non-optimized band offset, which results in lower open-circuit voltage (V_{OC}).

Energy levels. Another factor that affects V_{OC} is the gap between the acceptor's lowest unoccupied molecular orbital (LUMO) and the donor's highest occupied molecular orbital (HOMO) levels. To achieve a higher V_{OC}, a donor polymer with a lower HOMO level or an acceptor with a higher LUMO level must be used.

Carrier mobility. Low mobility of conjugated polymers results in poor charge transport and charge collection, lowering external quantum efficiency. Enhancing π–π^* stacking or the regularity of molecular structure within polymers can improve mobility.

Morphology. Higher molecular crystallinity can result in better π–π^* stacking, higher absorption, higher mobility, balanced charge transport and higher efficiency. A much better understanding of morphology is needed to achieve these results.

A unique property of PSCs is their potential for being semitransparent or translucent. A major limitation of active polymers, as discussed previously, is their limited absorption of the solar spectrum, partly because the polymer active-layer in the solar cells is usually very thin, in the range of 100–200 nm. As a result, a significant amount of light remains unabsorbed. Even at the peak absorption wavelength, the transmittance is still around 60–70%. This opens the possibility for the realization of translucent solar cells, composed of a very thin, polymer active-layer sandwiched between two transparent electrodes. The design for one type of translucent solar cell is shown in Fig. 2b. This type of cell does not rely on a grid- or mesh-type electrode (which has alternate opaque and transparent areas), but instead has electrodes that are completely transparent throughout the film surface, achieving transmittance as high as 85%.

An interesting aspect of this cell design is its reverse polarity. The bottom electrode, consisting of a TCO layer coated with very thin n-type interfacial materials, makes it a cathode for collecting electrons by lowering the work function. The top electrode — the anode — is formed by depositing a very thin layer of p-type interfacial material, followed by a transparent conductive film (which may be a TCO or metal such as gold). This is referred to as the 'inverted' cell configuration. By varying the thickness of the transparent electrodes, the transparency and the efficiency of the cell can be controlled. Transparency as high as 40% (on average) in the visible range, with a photovoltaic power-conversion efficiency of up to 2.5%, has been demonstrated. Furthermore, by choosing polymers with different absorption bands, the colour of the cells can be tuned (Fig. 3). The fabrication process for translucent solar cells is similar to that of regular PSCs; they could therefore be made using the same manufacturing line at similar high throughput and low cost.

PSCs aim to help address the most challenging issue of solar-cell technology — the high cost of manufacturing and materials. The realization of low-cost PSCs will help solar-cell technology to be used in many other applications. The first one is the consumer electronics market, where there is a clear need for built-in power generation to help support or supplement existing battery technology (which is struggling to keep pace with the power requirements of portable devices). PSCs are well-suited to this application owing to their flexibility, light weight, low power needs and the relatively low lifetime requirements of such devices. The first polymer solar panel products are likely to be chargers for portable electronic devices (such as mobile phones and personal digital assistants (PDAs)), and will be available to consumers in late 2010 or early 2011. Figure 1 shows prototypes where PSCs are integrated into products, such as a charging pad for a global positioning system (GPS) unit, or a portable power source integrated into a messenger bag.

The second area of application is in 'smart fabrics', where PSCs can be integrated into various textiles and fabrics for use in clothing, wallpaper, curtains, tents and other similar products. Figure 4 shows an artist's rendition of green-coloured PSCs integrated into an awning of a building. During the day, the solar cells convert sunlight into energy, which is stored in batteries. During the night, this energy can be used to provide lighting or to power other electrical appliances. Such applications are ideal for remote, off-grid locations where other electricity sources are not easily available. Furthermore, PSCs can be integrated into military uniforms to provide power for lighting, heating and remote command technology, while being lightweight and mobile. A big advantage

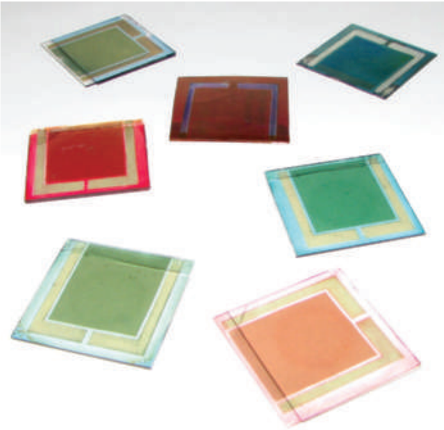
SOLARMER

Figure 3 | Translucent polymer solar cells of different colours and transparencies. The colour of the cells can be changed by using different polymers for the active layer. The transparency of the cells can also be tuned by controlling the thickness of the polymer active-layer and the electrode thickness. Such coloured translucent solar cells provide an added dimension for architects and designers for integration into commercial products.

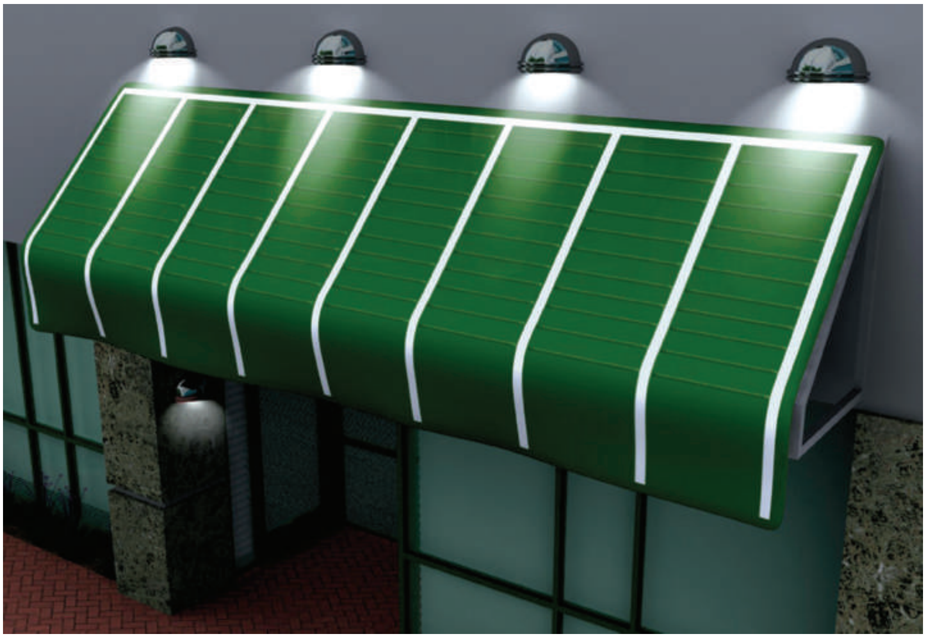
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Figure 4 | An artist's rendition of flexible polymer solar cells (PSCs) integrated into an awning. The cells absorb sunlight during the day and convert it into energy, which can be stored in batteries and used to power electrical appliances (for example, lighting) during the night. The ease of manufacturing large-area PSCs at relatively low cost will enable such applications in the near future.

of PSCs is that they do not require directly incident sunlight to generate power; they respond very well to indirect and low-light conditions, such as on a cloudy day. The market for smart fabrics is estimated to grow to $700 million by 2011, and PSCs have the potential to be a major part of this industry. These products will also provide power for outdoor and recreational sports, as well as for remote and off-grid power.

In the long-term, when PSC lifetimes and efficiencies have evolved towards that of traditional solar technologies, they may have application in the building integrated photovoltaics (BIPV) market, or other grid applications. This presents a tremendous opportunity because the solar market is estimated to grow to $74 billion by 2017. Potential applications in this area include roofing, facades, smart windows and walls. The major drivers for the use of PSCs in BIPV are flexibility, easy installation, good performance in dim light, aesthetic value for architecture, and low cost.

There are still several major challenges that must be overcome to realize the full commercial potential of PSCs. At present, maximum efficiencies are around 6% — a significant improvement from less than 1% about 5 years ago. However, for commercial applications such as the ones listed above, higher efficiencies are essential. Another challenge is that PSCs have relatively short lifetimes, currently around a few thousand hours of continuous operation under normal lighting conditions. For widespread market acceptance of this technology, product lifetimes of at least 3–5 years and efficiencies in the range of 8-10% must be achieved. Factors that affect the lifetime are temperature, cycling of temperature, humidity, relative humidity, light sources and cycling of light. The stability of packaging, substrates, electrodes, and load conditions also affect the lifetime of PSCs and as a result, advanced encapsulation technologies need to be developed to protect solar cells from these elements. The organic light-emitting diode (OLED) industry has made significant progress in dealing with lifetime issues for their devices, providing significant hope for the PSC industry. Many large companies such as Vitex Systems, Alcan Packaging, Toppan Form, Honeywell, DELO and others are developing barrier films for enhancing the lifetimes of PSCs for practical applications.

So far, the record efficiency values for PSCs have been demonstrated only for laboratory-scale cells; more work is required to scale up to manufacturing levels. Most of the potential benefits — low-cost, easy fabrication over large-areas, and flexibility — have yet to be demonstrated at production levels. Efficiencies of manufactured PSCs are expected to be lower than that of lab-scale cells, highlighting the need to develop a high-throughput printing process for manufacturing. The most significant advantage of this technology is the projected low cost, which will position PSCs further ahead than any other solar-cell technology already in the market or currently under development. To realize this cost advantage, manufacturing processes must be developed to enable production at around $50 m^{-2}. The PSC industry is clearly making progress: Plextronics has reported NREL-certified 2.05% module efficiency for a 6 × 6 square inch module made on glass; Konarka has produced prototypes of flexible solar panels from their pilot production line and has announced plans to establish a manufacturing line with a capacity of 1 GW; and Solarmer will move from lab prototypes to pilot-scale manufacturing in early 2010.

The focus for the future is mainly on two major fronts — to develop manufacturing processes that will enable production of PSCs at high throughput, high yield, and low cost; and to improve the lifetime of these cells to 10 years and beyond, so that they can compete with other solar technologies for large-scale power generation. ❒

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MATERIALS PROCESSING

Two-photon fabrication

Maria Farsari and Boris N. Chichkov

Two-photon polymerization is a 3D nanoscale manufacturing tool that offers great potential for rapid prototyping and the manufacture of photonic devices, tissue scaffolds and biomechanical parts.

The rapid development of femtosecond laser systems and their transformation from research to production tools is triggering the industrialization of laser applications based on nonlinear optical processes, which so far have been confined to the laboratory. Direct laser writing by two-photon polymerization (2PP) of photosensitive materials is an example of such an application, and has emerged as a promising technique for rapid and flexible fabrication of fully three-dimensional (3D) structures with sub-100-nm resolution. The technology is based on two-photon absorption; when the beam of an ultrafast (for example, femtosecond) laser is tightly focused on a volume of photosensitive material, the polymerization process can be initiated by two-photon absorption within the focal region. By moving the beam focus in a 3D manner through the material, 3D structures can be fabricated. The only processing required afterwards is the washing and removal of the non-illuminated — and therefore non-photopolymerized — material.

The attraction of 2PP technology is that it can create computer-designed, fully 3D structures with resolution beyond the diffraction limit — no other competing technology offers these advantages. Classic 3D prototyping techniques, such as UV laser stereolithography and 3D inkjet printing, can also reproduce fully 3D structures; however, they provide a maximum resolution of only a few micrometres. Lithographic techniques with superior resolution, such as e-beam lithography, are limited to producing high-aspect-ratio 2D structures.

Two-photon polymerization was first demonstrated in the 1990s — since then, a large number of research laboratories have been involved in its study. Despite the unique capabilities of 2PP and its potential applications in fields such as micro/nanophotonics, micro-electromechanical systems (MEMS), microfluidics, biomedical implants and microdevices, it has yet to be applied to manufacturing. This is due to a number of factors: for example, the relatively high costs of femtosecond lasers, positioning systems and optics; the slow speed and small volume of processing; and the lack of specially designed materials with good optical, mechanical and biomedical properties. However, many of these issues are being resolved, and 2PP is transforming from a research to a production tool.

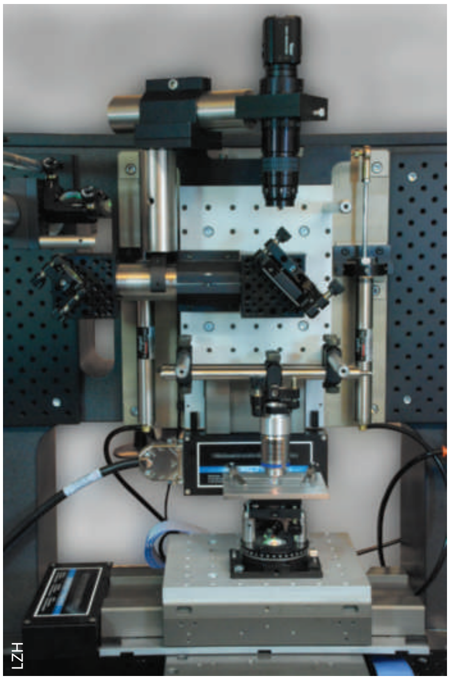

Figure 1 | Inside view of the front of the commercially available 2PP system (housing open). The laser beam is guided by a set of mirrors to the microscope objective. The sample is placed below the microscope objective on a small platform mounted on a nanopositioning system. The photopolymerization process can be monitored online using a camera (upper part of the picture).

In recent years, considerable progress has been made in improving the reliability and reducing the cost of femtosecond laser oscillators and laser systems. For 2PP, no specialized laser source is required, as off-the-shelf femtosecond laser oscillators (available from different laser suppliers) can be used. The use of femtosecond laser pulses is essential for initiating a nonlinear optical process, as high intensities in the focal volume are required. However, the overall laser power needed is low — a few milliwatts are sufficient if the laser pulses are focused with a lens having a high numerical aperture.

The cost of femtosecond laser oscillators has not been the only factor hindering the industrialization of 2PP; for example, another issue is the slow speed and small volume of fabrication. To benefit from the high resolution of this technology, very accurate positioning systems — traditionally piezoelectric stages with a small working area — must be used. Laser Zentrum Hannover (LZH) has designed and built an autonomous compact 2PP machine that integrates a femtosecond laser, a scanner for fast writing of small-area structures and a motor-driven linear positioning system (Fig. 1). The three-axis positioning system provides a travelling range of 10 cm in each direction and a processing speed of up to 30 mm s^{-1}. This autonomous 2PP machine is now commercially available.

Most of the materials used for 2PP are designed for conventional lithographic applications and there have been examples of both negative and positive photoresists being used. In the case of negative photoresists, the two-photon exposure results in the crosslinking of polymer chains, making the exposed area insoluble to the solvent used to remove the unpolymerized material — the structure is written directly in the sample. With positive photoresists, the opposite occurs; two-photon absorption causes the photoresist polymeric chains to break and become soluble to the development solvent, so the reverse structure is written in the sample. Negative photopolymers are most commonly used, with the most popular being the UV lithographic photoresist SU-8, and the hybrid sol–gel ORMOCER (Microresist Technologies). Both are commercially available materials that

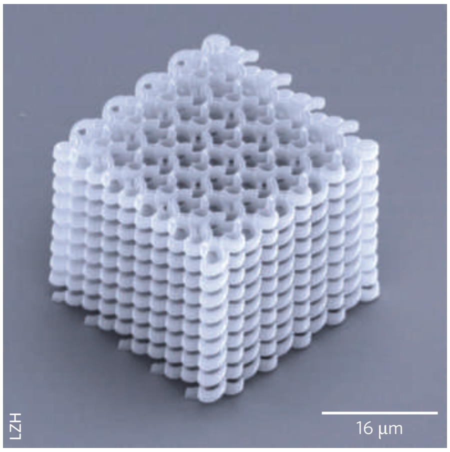

Figure 2 | A spiral photonic crystal fabricated using the zirconium/silicon hybrid sol–gel SZ2080. As the material does not shrink during photopolymerization, there is no need for extra support structures or frames.

give very good results for resolution and processing. The main disadvantage of all commercially available materials, however, is that they cannot be easily modified or combined with active components for added functionality. For example, the incorporation of organic molecules with nonlinear optical properties could be used for the fabrication of 3D nonlinear photonic devices, and metal-binding chemical groups could be useful for the fabrication of metamaterials.

LZH and the Foundation for Research and Technology Hellas (FORTH) have been collaborating on the development of new, organic/inorganic hybrid sol–gel materials, specifically designed for 2PP applications (Box 1). The technology behind sol–gel materials provides a powerful tool for the development of photosensitive compounds. These materials benefit from straightforward preparation, modification and processing, as well as having high optical quality, post-processing chemical and electrochemical inertness, and good mechanical and chemical stability. Thus, it is clear that these materials may have many applications in photonics, MEMS, microfluidics and biomedicine. The materials developed through the LZH and FORTH collaboration include highly transparent, biocompatible photopolymers that do not distort during polymerization, and active materials with nonlinear optical functionalities and metal-binding affinity.

The high resolution and 3D nature of the 2PP technique, combined with the high transparency of sol–gel materials, make it an ideal technology for the fabrication of nanophotonic devices. Of particular importance is its application in complicated 3D structures, such as photonic crystals. These are periodic structures considered to be the optical equivalent of semiconductors; they modify light in the same way a semiconductor does for electrons. 2PP is not the only technology for the fabrication of periodic 3D nanostructures; other methods include holographic interferometry and colloidal self-assembly. However, to be practical for photonic circuits, it is essential to remove symmetry and introduce defects into 3D photonic crystals — something not possible using other methods. An example of a photonic crystal made using 2PP is shown in Fig. 2. Here, the material used was the zirconium/silicon hybrid sol–gel SZ2080, developed by FORTH and LZH. One characteristic of this particular material is that it does not shrink or distort during structuring, making the design and fabrication of photonic devices much simpler and repeatable.

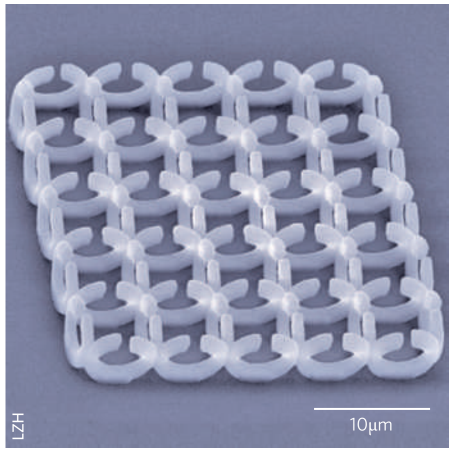

Figure 3 | A 3D split-ring metamaterial structure, fabricated using a metal-binding hybrid polymer composite. 2PP is the only technology that can be used for the fabrication of 3D free-standing structures with sub-micrometre resolution.

Photonic-crystal fabrication is not the only area of photonics in which 2PP has potential applications. So far, other areas that have been identified are micro-photonic components, such as micro-prism and micro-lens arrays, diffractive optical elements, and metamaterials. Metamaterials are artificial materials that, unlike naturally occurring substances, exhibit magnetism at optical frequencies; it is therefore essential for the application of 2PP that the structures are conducting. However, the fabrication of 3D metallic structures with sub-micrometre resolution is not an easy task — most of the metamaterials reported are 2D structures, fabricated using electron-beam lithography and evaporation of metal films. So far, the fabrication of high-quality metallic 3D structures using 2PP has not been achieved. A more feasible approach is to first fabricate dielectric structures, then metallize them in a second step. Two methods of post-fabrication metallization have been demonstrated: silver chemical vapour deposition and electroless-plating metallization. The first method is independent of the 2PP material used for the fabrication of the structure. The resolution and dimensional accuracy of the structures is therefore not affected, but the technique is limited in the number of different geometries that can be metallized. For the second method, a metal-binding photopolymer (or an appropriately functionalized surface) is used, and the metallization is done using

Box 1 | Sol–gel reaction

The sol–gel process is a relatively new material technology, and is proving to be a powerful tool for the fabrication of 3D structures from organic–inorganic hybrid materials. The process can be explained by four steps.

The first step is hydrolysis and condensation, where precursors or monomers (such as metal alkoxides) are mixed with water, and undergo hydrolysis and condensation to form a porous interconnected structure. Either an acid, such as hydrochloric acid (HCl), or a base, such as ammonia (NH_3), can be used as a catalyst. The second step is gelation, where the liquid solvent (acid or base) is removed and a gel is formed by heating at low temperature. It is at this stage that any significant loss in volume occurs. The third step of the process is photopolymerization — the use of light to trigger transformation from a gel to a hard material similar to glass. If a photoinitiator is present in the gel, the regions irradiated by light will photopolymerize, owing to the linkage of carbon–carbon double bonds in the organic material. At this step there is no material removal and no volume loss. The last step is the development: the sol–gel is immersed in an appropriate solvent and the area of the sol–gel that is still a gel — that is, material that was not photopolymerized — is removed.

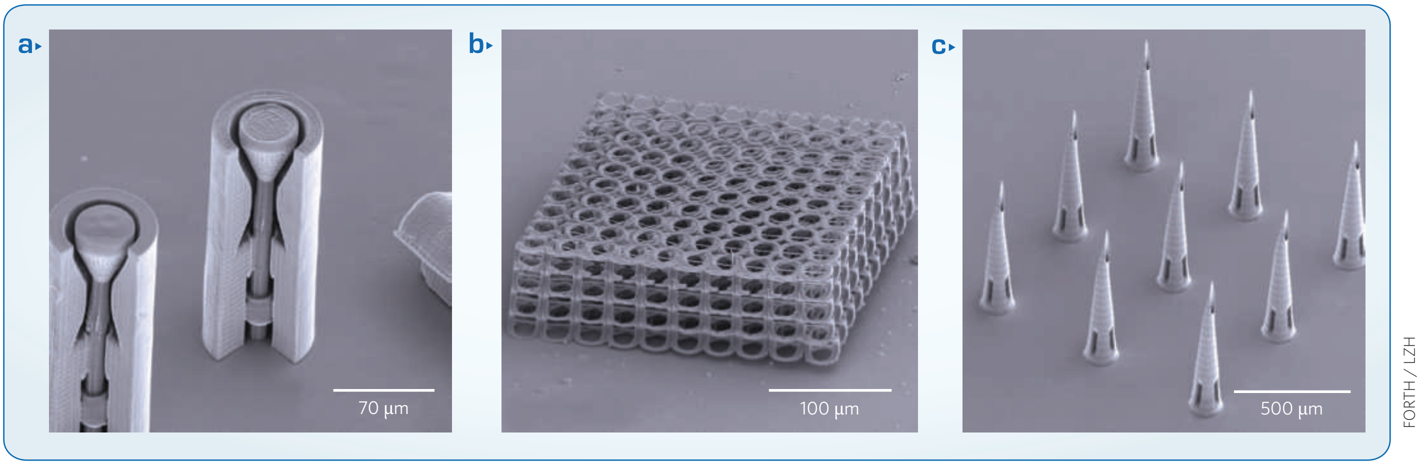

Figure 4 | A selection of biomechanical parts made by 2PP. **a**, Microvalve designed to prohibit the reversal of blood flow in human veins. Only part of the valve cover was built, to enable visualization of the interior. **b**, Scanning electron microscope image of high-porosity tissue engineering scaffold. **c**, Test micro-needle array for transdermal drug delivery.

silver electroless plating. So far, structures fabricated using this technique have not had the required resolution to operate as metamaterials because, in most cases, acrylate photopolymers have been used, and these suffer from distortion due to shrinkage during photopolymerization. The metal-binding hybrid materials developed by FORTH and LZH show considerable potential for both resolution and metallization capability (Fig. 3). Using these materials — which contain a metal-binding amine group — structures with a resolution down to 100 nm can be achieved. Furthermore, conductive atomic force microscopy studies have shown that the full metallization of the structure surface is realized.

Another area where the potential of 2PP technology is being realized is in biomedical applications. The biological response of a series of commercially available and in-house photopolymers has been investigated, and several were found to be biocompatible. This, combined with the ability to structure them in complicated 3D shapes, makes them attractive for applications such as implantable MEMS and scaffolds for tissue engineering. One such example of implantable MEMS is shown in Fig. 4a. This complex shape is a readily assembled check-microvalve, designed to prohibit the reversal of blood flow in human veins, which may be caused by standing for too long. The valve is designed to open under forward fluid-flow and close firmly in the case of backward flow. As 2PP is a direct-write technique, the dimensions of the valve can be changed by simply scaling the computer design to fit the patient's requirements, and its fabrication takes only a few minutes. It is made with SZ2080, the same zirconium/silicon hybrid material used for the fabrication of the spiral photonic crystals of Fig. 2. In this application it benefits from the dimensional accuracy and lack of shrinkage of this material, as well as its biocompatibility.

The ability to produce arbitrary 3D scaffold structures using biocompatible materials is also appealing for tissue engineering. These scaffolds are required in order to produce living tissue that can integrate with host tissue inside the body (Fig. 4b). Micrometre-sized topography has been shown to have an essential role in determining cell adhesion and surface-bound characteristics, influencing important cellular functions such as survival, proliferation, differentiation, migration or mediator release. In particular, 3D cell cultures offer a more realistic local environment where the properties of cells can be observed and manipulated. An important factor in the production of tissue-engineering scaffolds is the ability to reproducibly manufacture 3D nanostructures; a direct application of 2PP technology.

2PP technology can also be used in the fabrication of drug-delivery devices, such as micro-needle arrays — devices that enable transdermal delivery of different pharmacological substances (Fig. 4c). 2PP enables not only the miniaturization of such devices, but also allows the adoption of more than one needle design in a single array, therefore optimising the effect of geometry on mechanical and puncturing ability. The micro-needle arrays are currently undergoing clinical trials to determine their effectiveness and suitability.

Now that 2PP machines are commercially available, the technology has started to expand and industrial interest is growing. So far, LZH has already delivered several units to clients in Europe who have realized the enormous potential of this technology. Furthermore, a number of companies from the telecom and biomedical sectors have been carrying out feasibility studies to evaluate the capabilities of 2PP technology. As with any new technology, the industry has been reluctant to embrace it and accept it as a production tool. This has also been hindered by the fairly limited portfolio of materials specially developed for 2PP, and by the relatively slow processing speed of the technology. However, the number of specially designed materials available has increased, and there is also the potential of increasing the processing speed using multibeam parallel processing. As there is no other viable alternative to fully 3D nanomanufacturing, the expansion of 2PP as a production tool looks very promising. ❒

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LED TECHNOLOGY

Organic displays come of age

David Fyfe

Eighteen years after the development of the first polymer-based LED display, the technology has finally matured and polymer OLED televisions are just around the corner.

So much has changed since the first display based on polymer organic light-emitting diodes (P-OLEDs) was developed in 1991. Today, many mobile phones use OLEDs and, with major players such as Sony, Samsung, LG and Chi Mei investing in the technology, the OLED television is now coming to market. Although currently all commercial OLED products are based on small-molecule OLED technology, polymer-based OLED devices are rapidly catching up in terms of performance. The evolution of P-OLED technology has been made possible by advances in all areas of science and engineering relating to displays: materials science, device structure and manufacturing technology.

Researchers first saw light emission from conjugated polymers in 1989. However, efficiency and lifetimes were extremely low — efficiencies were only a fraction of a lumen per watt, and lifetimes were measured in minutes.

The initial discovery was based on polyphenylene vinylene (PPV), but in 1997 the emphasis shifted to another class of conjugated polymers — polyfluorene-based, which give a wider colour range of emission and enhanced stability. This material is still the core platform for P-OLEDs. The device structure incorporating the P-OLED light-emitting polymer layer (LEP) has also undergone substantial evolution. The constant struggle for developers is to achieve efficient charge injection, as well as to balance the flux of electrons from the cathode with that of holes from the anode. This is to ensure that electron–hole recombination in the LEP layer — the process by which light is generated — is optimal.

Efficiencies and lifetimes vary depending on the colour of the OLED emission. Lifetimes in particular vary with the brightness of the pixel, according to an approximately square power relationship; doubling the brightness reduces the lifetime by a factor of four.

Development of phosphorescent red LEPs has been so successful that development has been halted to allow focus on green and blue emitters (Box 1 Table B1). For red light emitters, lifetimes in excess of 200,000 h (from an initial brightness of 1,000 cd m^{-2}) and efficiencies in excess of 20 cd A^{-1} have been achieved — a performance that satisfies the requirements for large-screen TVs. The development of a green phosphorescent material with a lifetime in excess of 40,000 h and an efficiency of >40 cd A^{-1} has also been accomplished. International colour standards have been met for both red and green light emitters.

Blue devices, having the largest bandgap, are the biggest challenge. However, huge advances have been made in the lifetimes and efficiencies of these devices.

Box 1 | The changing market

Until 2005, there were essentially four companies developing LEP materials: Cambridge Display Technology (CDT), Dow Chemical, Sumitomo Chemical and Covion (then owned by Avecia of the UK). In 2002, CDT had licensed material intellectual property (but not device manufacture) rights to Dow, Covion and Sumitomo Chemical, believing that its licensing position would be enhanced by strong materials development. At the time, CDT's main materials research was on polyfluorene-based LEP materials, for which Dow held key patents.

Also in 2002, CDT bought the intellectual property rights of Opsys, a spin-off company from Oxford University in the UK. Opsys had developed a printable, phosphorescent small-molecule-based technology focusing on green colour. In this approach, a small-molecule emitter was sheathed in a dendrimer, stabilizing it when dissolved in a solvent for printing.

Universal Display Corporation was developing high-efficiency, phosphorescent, vacuum-processable small molecules and CDT was losing ground in the comparison of efficiencies. Furthermore, CDT had been making little progress with a high-efficiency, long-lifetime fluorescent red emitter. With the acquisition of Opsys, CDT (with the collaboration of, and funding from, Sumitomo Chemical) quickly developed a printable phosphorescent red using a phosphorescent (iridium-based) small-molecule emitter in a polyfluorene polymer host, and achieved huge advances in lifetime and efficiency (Table B1).

In 2005, German company Merck bought Covion, and Sumitomo Chemical bought Dow's P-OLED business, thus consolidating the intellectual property rights into two main competitors. Soon after, CDT and Sumitomo formed Sumation, a 50/50 joint venture. Thus, by 2006, the four P-OLED developers had become two — Merck with a spirofluorine intellectual property base (in addition to its older, more mature, PPV base) and Sumation with a polyfluorene intellectual property base. Since the 2007 merger of CDT into Sumitomo Chemical, research at Sumation has become an integral part of Sumitomo Chemical's P-OLED business.

Table B1

Colour	Colour coodinates	Lifetime, 2005 (h)	Lifetime, today (h)	Improvement
Red (phosphorescent)	(0.63, 0.37)	5,000	>200,000	40x
Green (fluorescent)	(0.31, 0.64)	4,000	115,000	30x
Blue (fluorescent)	(0.14, 0.18)	900	18,000	20x

LT_{50} from 1,000 cd/m^2

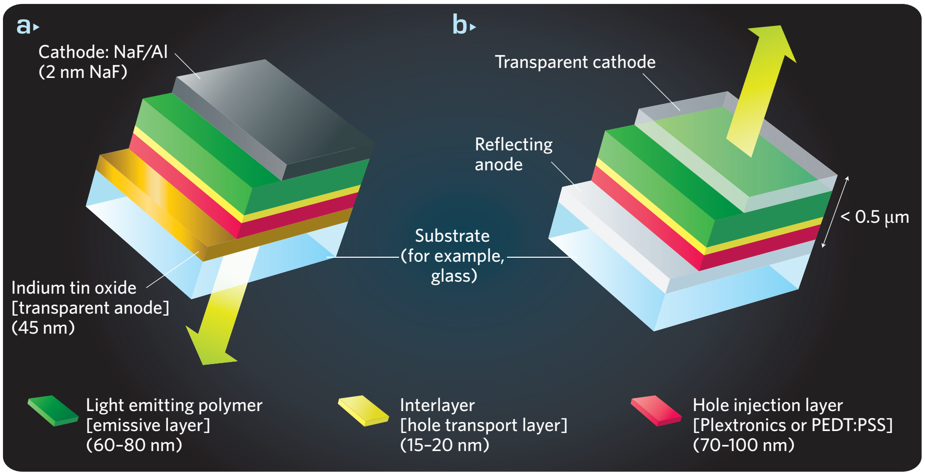

Figure 1 | Device structure of a P-OLED. **a**, Bottom-surface emission design. **b**, Top-surface emission design.

The current focus is on fluorescent blue with an improved colour point (a deeper hue). However, obtaining highly efficient, phosphorescent, deep blues with sufficient lifetime to meet TV requirements is a tough challenge — current fluorescent blue lifetimes (from 1,000 cd m^{-2}) are in excess of 30,000 h, and efficiency is about 8 cd A^{-1}. Lifetimes in blue are highly dependent on the colour point; a deeper blue gives a shorter lifetime.

The biggest advancement in P-OLED lifetimes and efficiencies followed the acquisition of the Dow Chemical P-OLED materials business by Sumitomo Chemical in late 2005, and the subsequent formation of Sumation (Box 1). The development substantially increased the library of monomers available to researchers.

There have also been important developments in the materials used on both the anode and cathode sides of a P-OLED device, and the way in which electrical charges are injected and transported into the light-emitting polymer. For many years, the transparent electrode material indium tin oxide — which has been commercialized for the LCD industry — has been the anode material of choice, despite evidence of indium migration into the hole transport layer.

In fact, most developments on the anode-side have focused on the hole-injection layer (HIL). Until recently, the material of choice for the HIL was poly(3,4-ethylenedioxythiophene):polystyrene sulphonic acid (PEDOT:PSS), a transparent conductive polymer mixture originally developed by Bayer. For a long time this water-based material provided the best device performance, but the component that provides strong conductivity, PSS, is a very strong acid, and consequently has not been a popular choice. Earlier this decade, it became clear that interactions between acid in the HIL and the emitting polymer layer were detracting from the efficiency and lifetime. In 2004, an interlayer (IL) or hole-injection layer was introduced into the device structure between the HIL and the LEP (Fig. 1), resulting in an almost sixtyfold increase in device lifetime and enhanced efficiency. Unfortunately, this added an extra processing step in P-OLED fabrication, but was deemed necessary because of the improved performance it brought about.

In recent years, other industry players have announced new HIL materials. Dupont's HIL is a PEDOT-based material, where the polymer that the PEDOT combines with was changed from PSS to Nafion, a Teflon-type material. Air Products and Plextronics have recently developed new self-doped HILs that offer significant benefits over PEDOT:PSS.

The IL layer has also been substantially improved. Although the LEP layer has remained at an optimum thickness of approximately 70 nm, the optimum thickness of the IL layer is substantially thinner (~15 nm). One challenge was to ensure that the solvent-based IL — once deposited on the water-based HIL layer — did not partially redissolve in the solvent of the LEP layer, reducing its thickness below the optimum amount. Crosslinkable ILs were developed, annealed at 180 C to ensure full crosslinking. The crosslinking chemistry used was selected to have no negative impact on device lifetime. The IL must be designed so that holes can be efficiently supplied into the LEP layer. However, to avoid hole–electron recombination — and thus light emission — in the IL (which would reduce the overall emission efficiency), it must be a wide-bandgap material. Thus, for every improvement made in the HIL or the LEP material, an improvement has to be made in the IL material.

Early cathodes were based on calcium with an aluminium capping layer. However, as the device structure evolved, lower-work-function cathodes became necessary and, in around 2000, a shift took place from calcium to barium. Inconsistent results led to another discovery — that barium oxide was an even better cathode than barium. In 2007, in an attempt to reduce drive voltage and thus lower power consumption, Cambridge Display Technology (CDT) changed to sodium fluoride cathodes, which resulted in further electron-injection efficiency, and lower drive voltage. It was found that to balance charge injection, a better hole injector (a better HIL) was required. The HIL that is currently preferred is made by Plextronics.

DEVICE STRUCTURE

In OLED devices where each pixel is powered by a transistor (so-called active matrix OLEDs), the light generated can escape either between the transistors (anode side or 'bottom emission') or, by making the cathode transparent, through the cathode ('top emission'). The advantage of the bottom-emitting device is that it is a simpler structure (Fig. 1a). The disadvantage is that the transistor blocks a significant proportion of the light and the pixels themselves have to be brighter to compensate; this ages the device more quickly and shortens the lifetime.

In the top-emitting structure (Fig. 1b), the proportion of generated light that reaches the viewing eye is significantly greater, enhancing efficiency and, because the pixel can be run less bright, extending lifetime. However, the device requires a cathode of high transmissivity and a reflecting anode (the bottom-emitting device has a reflecting cathode and a transparent anode).

At CDT we have had a focused top-emission device programme for the past two years, which has had considerable success. However, as with top-emitting small-molecule devices, the structure is significantly more complex than those of bottom-emitting devices. Therefore, we currently balance improved lifetime and efficiency of top-emitting structures with increased complexity, which will probably reduce manufacturing yields. It is not yet clear whether bottom emission or top emission will dominate when P-OLEDs enter large-scale manufacturing.

MANUFACTURING TECHNOLOGY

It has always been clear that the main advantage of P-OLEDs over vacuum-processed small molecules is in the printability of LEPs and associated layers in air. Today, despite breakthrough claims being made periodically by equipment makers, small-molecule device production under high-vacuum conditions using a perforated mask (shadow masking) to create the RGB pixel structure is stuck at 'half cut' processing of Gen 4 (730 mm × 920 mm) substrates. The substrate is processed by deposition through multiple masking steps to produce separate red, green and blue pixels in successive operations on half the substrate; the process is then repeated on the other half. In contrast, current LCD panels are now being made from Gen 10 (2,850 mm × 3,050 mm) substrates, highlighting the huge challenge that small-molecule technology has in becoming a mainstream contender for use in TV panels.

Kodak's approach to this challenge has been to implement a colour-filter-on-white approach, whereby a white OLED is vacuum-deposited on all pixels and red, green and blue sub-pixels are created — as in LCD panels — with a colour filter. Kodak's scheme allows some white light to pass through directly to enhance efficiency. However, there is still a 25% power penalty compared with an RGB pixel structure and many feel this is unacceptable.

With P-OLED manufacturing technology, the initial challenge was to develop a printer/printhead combination of sufficient accuracy and robustness. Seiko Epson was a pioneer in this area but preferred to keep its technology for internal use (although this policy may be changing). Printhead technology is continually improving, with drop sizes decreasing down to 1 picolitre (but more typically 6-8 picolitres) (Fig. 2). Furthermore, the angle of deviation of the droplet travelling from the print head to the substrate being printed is continually decreasing, owing

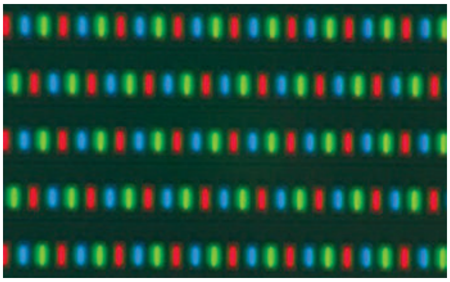

Figure 2 | Today's printing technology is able to accurately print pixels down to 1 picolitre. These pixels are 77 μm long and 32 μm wide. The pixel pitch — separation between pixel centres — is 53 μm horizontally by 159 μm vertically.

primarily to developments at Dimatix (acquired by Fuji Film) and Konica Minolta (and more recently by Xaar). However, in 2002, no companies were making a printer with the required stage precision and robustness for P-OLED printing. Recognizing the limitation this placed on adoption and commercialization of its core intellectual property, CDT purchased Litrex from Gretag of Switzerland in 2002 and funded development of commercial Gen 2 (370 mm × 470 mm) printers and a prototype Gen 7 (1,870 mm × 2,200 mm) printer. Two development Gen 4 printers were supplied to an Asian customer in 2005.

Although development was focused on printing P-OLED displays (Fig. 3), the traditional LCD industry became interested in the technology. As LCD substrate sizes reached Gen 5 (1,100 mm × 1,300 mm), interest heightened in printing alignment layers and spacers, as well as colour filters for Gen 7 and larger substrates. CDT, having proved that inkjet printing was feasible for P-OLED deposition on an industrial scale, sold Litrex to Ulvac (Japan) in 2005. Ulvac has subsequently built Gen 8 machines for colour filter printing.

This, however, was not the only printing challenge. When the first test OLED devices were printed, it was found that lifetime and efficiency were less than one third of that for the same material used to create a spin-coated device, and this applied to all three colours. In a spin-coated monochrome device, the P-OLED is applied at uniform thickness across the panel, and pixilation is affected by the cathode structure. In an RGB device, pixilation is affected by separating the P-OLED into red, green and blue sub-pixels and blanket coating the cathode.

The RGB pixels are created by first photo-patterning a 'black matrix' well structure. The dimensions of each well depend on the screen resolution required, and very accurate placement of the red, green and blue emitting polymers into the wells is required. This is also true for both HIL and IL materials, which are also printed. To facilitate this, a pre-deposition step of plasma treatment has been introduced. This creates hydrophilic wells with low contact angle, and well tops that are hydrophobic (high contact angle). Thus, any droplets that hit the top of the bank during printing are rejected by the surface and are drawn into the target well.

Each layer — HIL, IL and LEP — has to be of uniform thickness to achieve long life and high efficiency for the device, and substantial work has gone into formulating the inks for each layer to achieve this uniformity. The key has

Figure 3 | This 3-inch diagonal P-OLED display has a resolution of 302,400 pixels at a pixel density of 160 pixels per inch. It was made using inks from Sumation and high-resolution ink-jet printing.

been to use a combination of high- and low-boiling-point solvents to control the evaporative process.

Work continues to refine the ink formulation, print process and printing strategies. Significant progress has been made towards achieving the same performance characteristics as devices made using spin coating; parity is expected to be achieved sometime this year.

Progress in P-OLED display development has been helped by continuous improvement in printhead design. The printers themselves have improved such as to substantially reduce drop placement inaccuracies arising from movement of the printer head relative to the substrate being printed. It is now possible to foresee Gen 8 and larger substrate sizes being printed in one pass using many heads 'ganged' together.

Inkjet is not the only printing method being explored. Toppan Printing has made excellent progress with flexographic (roll) printing but, beyond Gen 5, issues related to flexing of the applying roller have still to be confronted. DuPont reports good results with nozzle printing, in partnership with Dai Nippon Screen.

We at Sumitomo Chemical/CDT believe that P-OLED is on the verge of becoming a commercially viable TV technology. Like any major technology shift, it has its champions and its detractors, but the move to OLED manufacturing by major players gives us strong belief that OLEDs will lead the TV industry in the foreseeable future. With the promise of displays with superior viewing angle, higher energy efficiency and lower cost to both manufacturer and consumer, we believe printed P-OLEDs will be the dominant OLED technology. ❐

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Large OLED displays ready for commercial production

SAMSUNG MOBILE DISPLAY

Samsung Mobile Display, which claims to be the world's largest manufacturer of organic light-emitting diode (OLED) displays, now has 14.1-inch and 31-inch OLED television panels ready for commercial production. The company claims that the 31-inch device features full high-definition resolution (1,920 × 1,080 pixels), a contrast ratio of 1,000,000:1, a colour gamut of over 100% NTSC and an ultra-slim design of only 8.9 mm. The OLED television panels can be mass-produced using fine metal mask technology.

The start date for commercial production has still to be decided. "We have not determined when the 14.1-inch and 31-inch OLEDs will be readied for production as that depends upon a number of factors, including our assessment of various market conditions and our readiness to invest," said John Lucas, public relations manager at Samsung Semiconductor.

The company has also developed a 'flapping' OLED panel — a panel that can flutter in a breeze. The panel is super-thin at only 0.05 mm, about one tenth the thickness of OLED panels with a normal glass substrate. It features a high contrast ratio, is polarizer-free and has a resolution of 480 × 272 pixels.

www.samsungsmd.com

Inkjet technology suitable for big-screen OLED televisions

The mass production of large-screen OLED televisions has been hindered by the lack of technology capable of depositing uniform organic layers on large substrates. However, Japan-based Seiko Epson Corporation has announced that its proprietary Micro Piezo inkjet technology can provide a solution to this problem.

Using the same drop-on-demand approach as an inkjet printer, Epson's technology is capable of depositing droplets of liquid organic materials accurately on a large substrate in the precise locations and amounts required. The technology resolves the uneven layering that had been an issue with previous inkjet methods, allowing formation of uniform layers with extremely low volume error (<1%) to be achieved. As it does not require a mask and uses materials extremely efficiently, the approach improves not only the quality but also the throughput of the deposition process. The company said that this represents a step forwards in realizing 37-inch-and-larger full high-definition OLED televisions.

www.epson.jp

Solar cells suit indoor use with artificial lighting

New Energy Technologies, a developer of alternative and renewable energy schemes based in Maryland, USA, has announced that its ultra-small solar cells are an effective solution for generating electricity from artificial light. At less than a quarter of the size of a grain of rice, they are said to be the smallest reported organic solar cells of their kind. The company is now working on combining the cells with transparent glass to fabricate a solar window that can generate electricity when illuminated with direct sunlight or artificial lighting, such as fluorescent tubes.

"One of the biggest issues with today's solar products is their dependency on direct sunlight, which our cells have demonstrated the potential capacity to overcome," explained Meetesh Patel, President and CEO of New Energy Technologies.

www.newenergytechnologiesinc.com

Integrated photovoltaic module exploits laser scribing

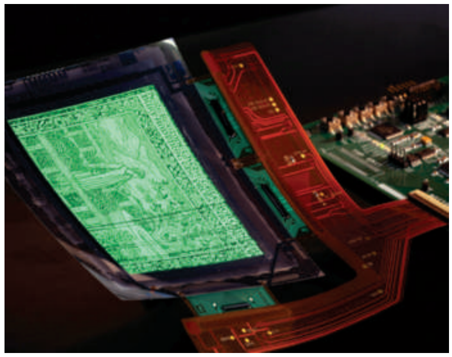
A highly integrated organic photovoltaics (OPV) module is the latest product of a collaboration between the Mitsubishi Corporation, the National Institute of Advanced Industrial Science and Technology (AIST) and the Tokki Corporation, all based in Japan.

OPVs can be used effectively in a wide variety of situations — windows, walls, cloths, textiles, outdoor equipment and toys — applications that have proven difficult for current bulkier silicon-type photovoltaic modules. OPVs are, however, inefficient in converting sunlight to energy. Now, the highly integrated technology developed by the Mitsubishi Corporation, AIST and the Tokki Corporation looks set to solve this problem.

The OPV module is based on technology developed by AIST in January 2005 that has an efficiency of 4%. It uses laser-scribing technology on a glass substrate, in which organic semiconductor materials are deposited and then divided into several cells. The technology eliminates the need for deposition mask patterning that is used in conventional methods. Being highly integrated, the new module is able to improve sunlight-conversion efficiency.

www.mitsubishicorp.com

A thin and flexible answer to active-matrix displays



FLEXIBLE DISPLAY CENTER

The Flexible Display Center (FDC) at Arizona State University and Universal Display Corporation (UDC) have worked together to develop an active-matrix flexible OLED display. The display is manufactured directly on DuPont Teijin's polyethylene naphthalate substrate and uses amorphous-silicon drive circuitry.

Implementing UDC's phosphorescent OLED (PHOLED) technology and the FDC's proprietary bond-debond manufacturing technology, the 4.1-inch monochrome quarter video graphics array display represents a significant milestone towards achieving a flexible OLED that suits mass production.

The flexible backplane display was manufactured at the FDC utilizing a 180-°C thin-film transistor process. The PHOLED materials allow the OLED to convert up to 100% of the electrical energy into light, as opposed to traditional fluorescent OLEDs which convert only 25%, providing up to four times more energy efficiency.

http://flexdisplay.asu.edu

The organic era

The organic photonics industry has come of age in the past few years. **Nadya Anscombe** speaks to Marc Baldo from the Massachusetts Institute of Technology, USA, about the advances that have been made and the challenges that remain.

Huge progress has been made in the last ten years in organic light-emitting diode (OLED) technology. What do you think are the most significant breakthroughs?

As a community, we have solved the internal efficiency problem using phosphorescent dyes and we are making progress with the stability issue. The stability and efficiency of blue OLEDs could still be improved, but for many applications the current performance is sufficient. Although great improvements have been made in the internal efficiency, there is still room for improvement in the external efficiency of OLED devices. Innovations in the optical design of OLEDs, such as texturing the transparent substrate, can decrease the amount of light that is trapped in waveguide and surface plasmon modes.

What are the main challenges that are still to be overcome in the OLED displays industry?

The biggest challenge faced by the OLED display industry is only indirectly related to OLED materials themselves. It is, surprisingly, the stability of the silicon backplane. If someone had told me back in 1997 — when I first became involved with this technology — that the stability of the silicon would be the problem in the future, I'd have laughed. Unlike other display technologies, the emission from an OLED is proportional to the current injected, making them especially sensitive to degradation in the backplane transistors that switch them on and off. Consequently, active-matrix OLED displays need especially reliable and stable transistors to drive them. This has meant that display manufacturers have had to use polycrystalline silicon for backplanes instead of less expensive amorphous silicon. Research is going on to try and solve this problem and companies are trying to develop new materials or use annealing techniques, but the problem remains. Another challenge is patterning a display, or defining the pixels. Most of the industry currently uses shadow masks, which are fine for small displays but are difficult to scale up to larger displays. When they are large they flex and also generate dust. Both of these are manufacturing issues. Although material development continues, most device issues have largely been solved — it is the manufacturing issues that now need attention.

MARC BALDO

Baldo: "Achieving an electrically pumped organic laser is a daunting technical challenge."

What trends are you seeing in emerging OLED products?

There are already a lot of mobile phones using OLEDs, but for larger displays manufacturing problems are expensive to solve. This is why it is the big companies that are coming out with products first. For example, Sony has launched an OLED TV and it really does look beautiful. However, with a price tag of a few thousand dollars, these are more of a statement of technical and financial resources than a mass-market product at the moment. When OLED displays were first launched, one of the unique selling points was the possibility of flexible displays. Again, however, the problem is in the backplane, and to a lesser extent, the packaging. The display industry will probably not pursue flexible devices commercially until an economical solution is found for the backplane in conventional displays. With all the manufacturing issues that OLED displays are facing, many companies have instead turned their attention back to the idea of lighting applications — particularly white light. OLEDs are ideal for large-area lighting applications and the big attraction of these applications is that they do not need patterning or an active matrix backplane — the two major issues in OLED display manufacture. So we are seeing major activities in OLED lighting, with some interesting products coming to market.

What is happening in the organic photovoltaics industry? Is it as mature and close-to-market as the OLED industry?

The organic photovoltaics (PV) industry has also made huge advances over the last ten years, but it is behind the OLED industry in maturity. Although it does not suffer from the same manufacturing issues as the OLED industry, stability of the organic compounds is potentially a bigger issue because of the exposure to solar radiation. The industry also cannot compete with conventional technology on material costs alone. It is not enough to say that organic cells are cheaper to manufacture; even if a cell is cheap, if it is not efficient, this will be offset by installation costs and the customer will not make any savings. Competing technologies such as cadmium telluride (CdTe) are now achieving costs of \$1 per watt and their materials costs are estimated to be less than \$0.05 per watt, which doesn't leave much room for organics. Efficiency is of utmost importance and there has been good progress made in this area, particularly with polymer blends. By incorporating fullerenes into organic photovoltaic cells, efficiencies of over 6% have been achieved. This is a dramatic improvement and very exciting.

What about other organic photonics applications, such as the organic laser?

Achieving an electrically pumped organic laser is a daunting technical challenge, but it isn't clear that it would solve any important commercial problems. Perhaps in the area of integrated photonics or green lasers, which are difficult to achieve using conventional materials, there may be scope for an organic laser. Of course, research into organic lasers may help to solve many issues that plague the OLEDs and PV industries, so research such as this is important.

INTERVIEW BY NADYA ANSCOMBE

Nadya Anscombe is a freelance science and technology journalist based in the United Kingdom.