

The World of Large-Area Glazing and Displays

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ABSTRACT

This work covers advances in large-area switchable glazing and flat panel displays. Large flat panels and glazing are being developed by a number of company and university groups. Certain novel flat panels display are made for electronic paper applications. Switchable glazing offers a new way of visualizing the function of a window. Switchable glazing can have a range of adjustable visible properties and shading coefficients. Technologies covered for glazing are electrochromism, suspended particles, encapsulated liquid crystals. Technologies being developed for electronic paper and certain flat display panels include electrophoretics, liquid crystals and bichromal balls. Beyond glazing applications, products based on this technology are flexible displays, electronic paper, switchable modulators, mirrors, and eyeglasses. This study covers developments from several companies including the one square meter electrochromic glazing made by Pilkington/Flabeg and Asahi/Nippon Mitsubishi Oil.

1. INTRODUCTION

This review is concerned with transparent glazing and large-area display media, especially media that can be made transparent in one switched state. Covered in this study are glazing and mirror technology based on electrochromics, dispersed liquid crystals and suspended particles. Also, a portion of the study is concerned with display media, including large flexible displays for such applications as electronic paper and selected flat panels.

The field of large-area smart media has been named Chromogenics^{1,2}. Chromogenics have some unique properties that have not been seen in other products for glazings. Electrochromics are favored for many applications because when they switch they remain specular, and non-scattering. This means they can be used for a variety of transparent glazing or see-through applications. Electrochromics can be easily powered because of their low voltage. Batteries or solar cells can power electrochromic glazing. Encapsulated liquid crystals (PDLC's) can not be used for this unless the application tolerates this, like upper windows and skylights. But, PDLC's are marketed on their unique privacy properties for security windows, utility vehicle side windows, and office privacy dividers. Suspended particle device (SPD) technology is similar to the PDLC's in that they are more absorbing or scattering in the off-state compared to the on-state. However, the SPD has an advantage of having much lower scattering in the off-state compared to PDLC's. Also, SPD's can be made to have a neutral color.

Commercialized chromogenic products fall into several different applications. They are medium-scale (>1 sq. m) architectural windows (Pilkington/Flabeg(UK/Germany)), electrochromics for automotive mirrors (Gentex (Zeeland, MI), Donnelly (Holland, MI), Toyota (Japan), Nikon (Japan), Murakami-Kaimedo (Japan)), PDLCs for privacy glass (Raychem (Mountain View, CA), Nippon Sheet Glass (Japan), Polyvision (Richardson, TX), 3M/Viracon (St. Paul, MN), Marvin Windows (USA), PDLCs for displays (Raychem, Polyvision), PDLC panels for large vehicle side glazing (GMC (MI), Ford (MI), and 3M). Other applications are electrochromic charge indicators (Eveready), electrochromics for sunglasses (Nikon) and electrochromic displays in cameras (Nikon). In the near-future, we expect to see the following products: electrochromic vehicle sunroof, SPD vehicle sunroof, SPD vehicle mirror, small skylights, switchable eyewear, SPD and electrochromic visors, banner displays, and aircraft side windows. Even in the more distant future, we expect to see large-area architectural windows, vehicle glazing, electronic paper and displays.

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2. ARCHITECTURAL AND VEHICLE MARKETS

Architectural applications have dominated the research and development of switchable windows. The flat glass market for architectural glazing is one the most attractive. There are wide possible applications in a variety of buildings. Also, specialty flat glass goes into a variety of products including flat panel displays. The use of flat glass is very wide spread, the world production of flat glass is about 2.4 billion m² per year (estimated for 2000), for all markets, with the largest portion going to building and automotive glazing. Window production in the U.S. alone is about 300 million m²/yr. The types of switchable glazing products for buildings are windows, residential insulated glass units (IGU), skylights, inclined glazing, security windows, commercial glazing, sunroom glazing, specialty glazing, doors, and stained glass glazing. However, the only largely successful switchable product has been mirrors for cars. The major drawback in the development of switchable windows is their size and lifetime requirements of 10-30 years. Several companies have undertaken the development of switchable windows. Pilkington/Flabeg (UK/Germany) have introduced their E-Control switchable glazing in Europe. Pilkington/Flabeg will install about 200 E-Control panels in a bank building in Dresden, Germany in 1999.

Chromogenic technology has a very important place in future vehicle glazing; it is one of the most exciting topics in window technology. Switchable technology can change how an automotive owner views a sunroof, side and back glazing, tint band, visor, mirror and information display system. Chromogenic technology can make an interior very comfortable and adjustable according to the driver's needs and changing visual environment. This technology can provide greater safety by virtual elimination of glare and reflections, which can fatigue or frustrate the driver. Dynamic glazing systems can be automatic, semi-automatic or have manual switching capabilities. The demand for switchable sunroof glazing is growing. Commercial electrochromic mirrors can be found on all major makes of cars. The chief maker is Gentex (Zeeland, MI). Other makers are Donnelly (Holland, MI) and Nikon (Japan) licensees. Prototypes of electrochromic automotive glazing panels have been shown by Donnelly, Flachglas (Germany) and Saint-Gobain (France), and Central Glass (Japan). Prototype automotive sunroof SPD panels have been made by Hankuk (Korea), a licensee of Research Frontiers Inc. (RFI). (Woodbury, NY) ^{3,4}.

3. DISPLAY MARKETS

The world market for displays is estimated to be about 38 billion US\$ in 1998. The display market is divided into two major technologies, the cathode ray tube (CRT) display and liquid crystal displays. The liquid crystal displays are based on both active and passive matrix designs. These displays are seen in a variety of electronic products ranging from televisions to palm top computers. Within the category of flat panels there are many new types of displays being developed. The US Display Consortium (USDC) supports some of these technologies. These new displays are plasma panels (PDP), Switchable glazing can also be used for larger information displays in applications where fast switching speed is not required, such as for airport display boards. There are three commercial electrochromic displays. Eveready makes one display for a battery tester. Currently, Nikon makes a display for SLR indicators in cameras. Minolta has made stock market board displays. Monsanto (St. Louis, MO) has developed a screen-printed electrochromic display on flexible plastic. The display is based on antimony-tin oxide. This technology could be used for banner displays ⁵.

A future display application is electronic paper, where information could be stored and displayed on a flexible substrate. Electrophoretic displays are being developed by E-ink (Cambridge, MA) for that application. In the E-ink display electrophoretic inks containing white particles are encapsulated in polymer bubbles and then embedded in a flexible polymer matrix. Depending on the applied field the white particles move up or down in the bubble giving a contrast effect. These displays operate at 90 V d.c and have contrast ratios of at least 6:1 ⁶. Also, they are bistable and exhibit a memory effect. Another electrophoretic emulsion development for electronic paper is being developed at NOK Corp. (Ibaraki, Japan) ⁷. A reverse emulsion nanosize electrophoretic display is being developed at Zikon Corp. (Campbell, CA). The Zikon display can appear transparent in one state then under applied potential change to reflective by particle aggregation⁸. Xerox PARC labs (Palo Alto, CA) is developing a flexible bichromal ball display sheet called the "Gyricon". This display is the result of many years of research. The Gyricon display switches as microballs rotate under applied field showing either light or dark pixels^{9,10}.

Some thin film display technology is directed at improving the quality of cathode ray tubes (CRT). A flexible display sheet using ferroelectrics is under development for CRTs by Idemitsu Kosan (Chiba, Japan). This sheet is used to rapidly switch the polarization of light for a 3-d television application. This sheet operates at 15 V d.c. with 1 mS response

time¹¹. Phillips has made a prototype electrochromic contrast enhancement filter for the face of a CRT. They were able to show a 75-15% visible change with 50,000 cycle lifetime¹². Also, both Donnelly and Saint-Gobain have developed electrochromic contrast enhancement panels for CRTs.

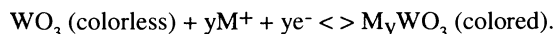
3. AEROSPACE MARKETS

The aerospace field is interested in the development of visors and windows that can control glare for pilots and passengers. Early work was done by PPG in the Flat Glass Division on electrochromic cockpit side windows. Several glazings were shown at the Paris air shows, but the product was cancelled a few years ago. In work on aircraft glazing, Dornier part of the Daimler-Chrysler Aerospace group (Germany), has been making prototype glazings for aircraft cockpits. They have shown a 0.3 x 0.3 m size laminated tungsten oxide/ polyaniline electrochromic window. Daimler-Chrysler Aerospace has announced dimmable windows for their first class section of the new Airbuss model A3XX airplane to be on the market in about 2002. Saint-Gobain has shown electrochromic prototypes of these new windows. They have a 40:1 contrast ratio with 1% transmission in their fully colored state¹³.

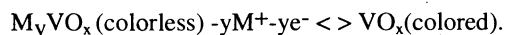
4. ELECTROCHROMICS

4.1 Electrochromic Materials and Device Design

Electrochromic devices are the most popular technology of large-area switching devices¹⁵. Much of the electrochromic technology is being developed for building windows and automotive mirrors and windows. Over 2000 patents have been granted on electrochromics. The major advantages of electrochromic materials are: (1) they have a small switching voltage (1-5 V); (2) they only require power during switching; (3) are always specular; (4) have gray scale; (5) have low polarization, (6) many designs have a long-term memory (12-48 h). Typical electrochromic glazing devices have upper visible transmission of $T_v=70-50\%$ and fully colored transmittance of $T_v=25-10\%$ and lower. The shading coefficient (SC) for electrochromic glazing is about $SC=0.67-0.60$ for the bleached condition, and $SC=0.30-0.18$ for the fully colored state. Electrochromic materials change their optical properties due to the action of an electric field and can be changed back to the original state by a field reversal. There are two major categories of electrochromic materials: transition metal oxides including intercalated compounds, and organic compounds (including polymers). The electrochromic effect occurs in inorganic compounds by dual injection (cathodic) or ejection (anodic) of ions (M^+) and electrons (e^-). A typical reaction for a cathodic coloring material using lithium as a coloration ion is:



A typical complimentary anodic reaction is:



These reactions show by using two different materials, composed of one layer that colors upon intercalation and one that colors on deintercalation, both sides of the device can color at the same time giving high optical density. Depending on the electrochromic, various coloration ions (M) can be used, such as: Li^+ , H^+ , Na^+ , and Ag^+ . The materials used in most devices are inorganic electrochromics: WO_3 , NiO , MoO_3 , and IrO_x ¹⁶. An electrochromic device must use an ion-containing material (electrolyte) bonded to the electrochromic layer as well as transparent conductors for setting up a distributed electric field. Devices are designed so they shuttle ions back and forth into the electrochromic layer with applied potential. An electrochromic window can be fabricated from three to five (or more) layers. A typical five layer device consists of an electrochromic layer, two transparent conductors, electrolyte or ion conductor, and an ion storage layer. Depending on the materials used in devices, some of the layers can be combined, serving dual functions. The most promising ion conductors are certain immobile solvent polymer systems, ionic glasses and open channel metal oxide structures such as Perovskites¹⁷.

Highly crystalline forms of WO_3 can have substantial near-infrared modulation. This material has the potential to control part of the infrared portion of the solar spectrum. The more common amorphous form, used by most developers, has only a little effect in the near-infrared region. Peak near-infrared switching levels from 20% to greater than 75% has been achieved with crystalline tungsten oxide¹⁷. The reflectance properties of tungsten oxide films produced so far seem to lie far from the theoretically limiting behavior.

Organic electrochromics are based on the viologens, anthraquinones, diphthalocyanines, and tetrathia-fulvalenes¹⁸. With organic compounds, coloration is achieved by an oxidation-reduction reaction, which may be coupled to a chemical reaction. The viologens are the most studied and used commercially of the organic electrochromics. Originally, organic electrochromics tended to suffer from problems with secondary reactions during switching, but more stable organic systems have been developed. The Gentex Corporation has commercially developed liquid organic electrochromic materials for automotive mirrors. Also, they have some effort to develop glazings. There were projects in the 1980's at Sony and Sharp in Japan and at IBM to develop a multicolor flat panel display using organic electrochromics. Uniform films of polythiophene, polyaniline and polypyrrole have been deposited by an oxidation polymerization technique. Polyaniline is the most favored organic polymer electrochromics.

4.2 Electrochromic Devices

Electrochromic mirrors are designed to automatically regulate glare in response to incident light levels. Both the Gentex Co. and Donnelly Co. have commercially developed electrochromic materials for automotive mirrors. A truck mirror commercialized by Donnelly is based on inorganic $\text{H}_y\text{WO}_x/\text{Ta}_2\text{O}_5/\text{NiO}_x$. Mirrors are the most commercially developed electrochromic products to date. Gentex has produced over 6.8 million mirrors over the last few years (1994-1998)¹⁹. The UV levels and upper temperature requirements are high for automotive sunroofs (95-100 C) making this application challenging.

The International Energy Agency (IEA) has investigated electrochromic materials and devices for building windows. There are several European government-funded programs. One of the bigger multinational projects is the Joule Commission of the European Communities (CEC) project. This project finished in 1998. Pilkington PLC (UK) had a multiyear project electrochromic glazing under the JOULE II program. This project involved several organizations, including Flachglas/Pilkington Deutschland and Davionics AS (Denmark), Oxford Brookes University and the University of Southampton. Pilkington/Flabeg has made several prototype "E-Control" switchable glazing of 0.80 m x 1.6 m. installed in a building. Panels up to 2 sq. m can be made. For the E-control window they obtained a range of visible transission of $T_v=50-15\%$ ^{20,21}. The glazing takes a few minutes to change color and to bleach.

Participants in the Eureka EC project include the Granqvist group at the University of Uppsala (Sweden) working with Coat AB (Sweden) and Leybold AG (Germany). In France, St. Gobain, Corning Europe, and C.S.T.B and the University of Domaine are working with electrochromic devices and components. In Italy, there are several groups involved in electrochromic devices testing and development. In Japan, Asahi Glass and Nippon Mitsubishi Oil have been steadily developing electrochromic windows of 1 sq. m based on $\text{Li}_x\text{WO}_3/\text{Li-polymer/Carbon}$ stripes for testing and evaluation. This glazing has transmittance, $T(633 \text{ nm}) = 60-19\%$ ^{22,23}. The Japanese Government's, Sunshine project, which funded part of Asahi's past work. The project goals were to develop a glazing with 50 % visible change; 10 year projected lifetime and high cycle lifetime. For this glazing Asahi Glass has obtained visible transmittance of $T_v=73-18\%$ and solar tranmittance of $T_s=55-11\%$ withstanding 100,000 cycles at 60 C²⁴. Some projected costs for electrochromic glazing are from 100-250 US\$/sq. m.

The nanocell photovoltaic was developed under the Prof. Gratzel group at Ecole Polytechnique (Switzerland). This is a new type of electrochemical photovoltaic cell. The cell relies on a active dye sensitized titanium oxide layer. Related to the nanocell is the photoelectrochromic device (also, known as a type of photochromic) where one of the electrodes is an electrochromic. The device can be designed to color in sunlight and regulate its color by shunting the anode and cathode of the cell through a variable resistor. The Prof. Gratzel group, Donnelly^{24,25} and NREL all have developed versions of this type of photoelectrochromic.

Sustainable Technology Inc. (STI) (Australia) is developing electrochromic windows with the help of the National Government Dept of Energy (ERDC). They have research partnerships with University of Technology-Sydney, Dept. of Physics also with the Monash University (Australia). STI used solgel deposition to produce their films on glass. STI uses a lithium salt loaded polymer for their ion conductor.

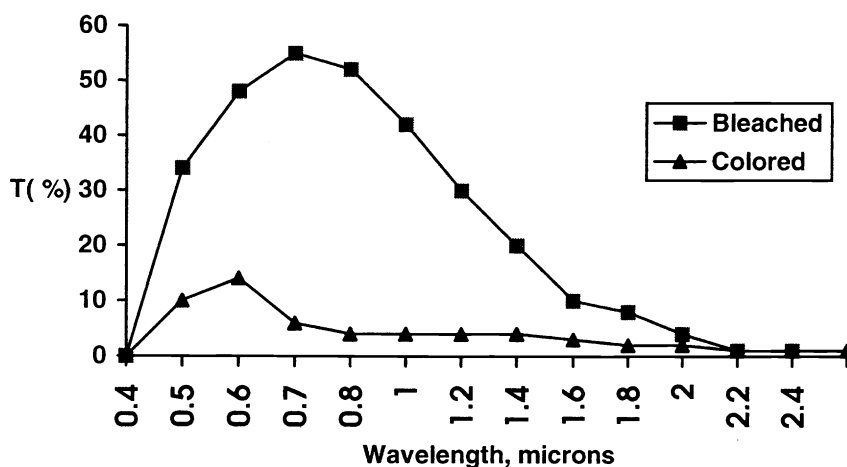


Fig. 4.1 Transmittance of an E-control electrochromic IGU made by Pilkington/Flabeg. Both the bleached and highly colored states are shown. (Source:Pilkington-Deutschland).

Under the 1995-98 U.S. DOE (Dept. of Energy) Electrochromics Initiative, both Donnelly Corp., and OCLI (Santa Rosa, CA) were awarded contracts to develop large-area electrochromic glazing. In 1998 a contract was awarded to Eclipse Energy (Gainesville, FL) for the development of PECVD electrochromics using the National Renewable Energy Labs (Golden, CO) (NREL) patents. Also, under the U.S. Dept. of Commerce, NIST grant, the SAGE Corp. (Faribault, MN) was awarded a sizable grant to develop electrochromics on glass. SAGE has formed a cooperative venture with Apogee Enterprises (Faribault, MN) to develop electrochromic glazing. NREL has been given the job of evaluating the lifetime and durability of electrochromic devices for the US National Department of Energy Program ²⁶.

5. LIQUID CRYSTALS

Liquid crystals account for the largest area of flat panel display technology. Liquid crystal display are rapidly improving in their properties and lowering in overall cost for flat panels. For display device industry is moving toward 1.1 m² glass sheets of 0.5 to 0.1 mm thickness as process standards for displays. In certain ways we are beginning to see a bridge forming between processing of large area glazing and large-area display glass. Some types of liquid crystals are nematic, smectic, twisted nematic, cholesteric, guest-host, and ferroelectric. For displays twisted nematic liquid crystal are by far the most commonly used liquid crystal. The mechanism of optical switching in liquid crystals is to change the orientation or twist of liquid crystal molecules interspersed between two conductive electrodes with an applied electric field. The orientation of the liquid crystal can alter the overall optical reflectivity properties of the window or display.

One unusual version of a liquid crystal system is an emulsion form of a liquid crystal and polymer to form a film. This emulsion is called phase dispersed liquid crystals (PDLC)²⁷ or nematic curvilinear aligned phase (NCAP)^{28,29} has been commercialized for use in switchable glazings. There are some preparation and structural differences between PDLC and NCAP, but here they will be treated without distinction since their performance is similar. The liquid crystals are encapsulated within an index matched polymer matrix. The polymer emulsion is fabricated between two sheets of transparent conductor coated polyester or glass that serves as electrodes. The switching effect of this device spans the entire solar spectrum, up to the absorption edge of glass. In the off-state, the device appears translucent white. When an electric field is applied, the liquid crystal droplets align with the field and the device becomes transparent. Typically these devices operate between 60-120 V a. c. (potentially as low as 30V). Their power consumption is less than 20 W/m² but require continuous power to be clear. In general, compared to electrochromics, the power consumption is higher for liquid crystals because of the need for continuous power in the activated state. The typical integrated hemispherical visible transmission values for a PDLC device are T_v (off-on)=50-80%. The shading coefficient changes by SC=0.63-0.79. Pleochroic dyes can be added to darken the device in the off-state. The dyed film shows considerable control over visible transmittance compared to an undyed film. The transmittance of both dyed and undyed films are shown in Figs. 5.1 and 5.2.

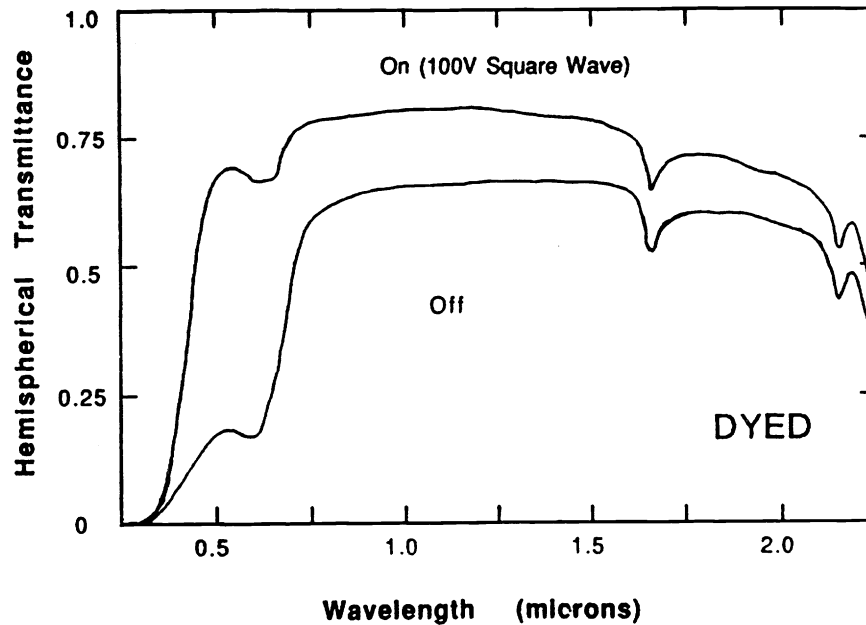


Fig. 5.1 Normal-hemispherical transmittance of an undyed PDLC film. (Source: Raychem)

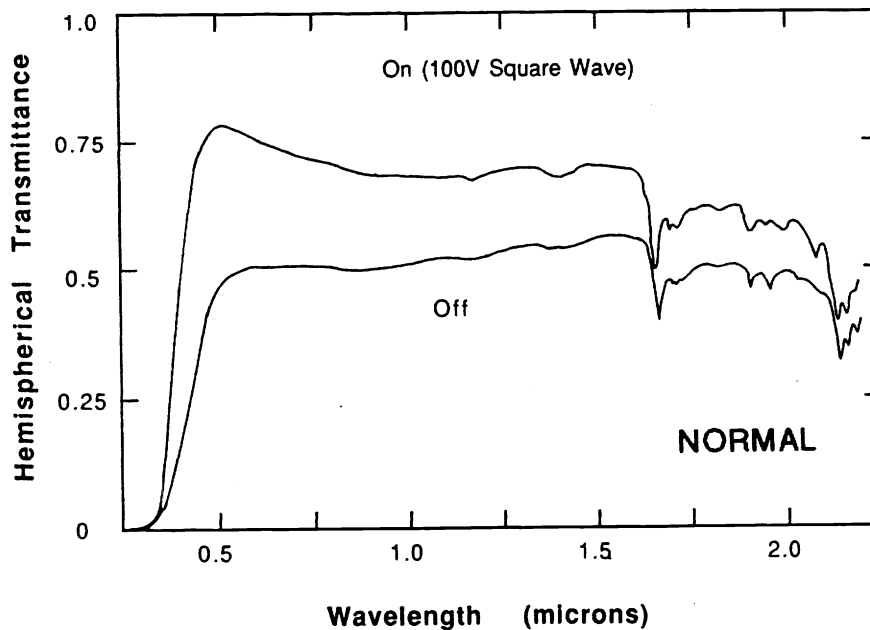


Fig. 5.2 Normal-hemispherical transmittance of a dyed PDLC film. (Source: Raychem)

In prior years, 3M had produced PDLC glazing panels for Marvin Windows (Warroad, MN) and Viracon (Faribault, MN)³⁰. Raychem (Sunnyvale, CA) licenses NCAP processes to Isoclima (Italy), St. Gobain and Nippon Sheet Glass (NSG, Tokyo, Japan). NSG produces an NCAP product known as "Umu" for specialty automotive and building applications³¹. Large-area NCAP glazing can be fabricated in 1 m x 2.5 m sheets. Open circuit memory is not possible with dispersed liquid crystals, without added dipoles to sustain the particle orientation. NEC and Toyota labs have developed a method to sustain the liquid crystal orientation after the external field is removed. They accomplish this by the introduction of a dipole particle that retains its local field. The device requires two separate frequencies to switch on or off³². By this technique Toyota has achieved 20 hours of memory. The Rohm and Haas company has made

improvements in PDLCs. They have lowered the switching voltage and reduced off-angle haze³³. Some of the issues that remain are UV stability and cost, which is about 750-950 US\$/sq. m. for glazings. A new type of liquid crystal panel has been shown by CLCEO (Santa Clara, CA). The panel is based on cholesteric liquid crystals. The panel shows very little haze when it is in the activated state, unlike the PDLC. The stability and durability properties of this panel have not been disclosed yet.

6. SUSPENDED PARTICLE DEVICES

The development of suspended particle or electrophoretic devices and glazing has spanned many years. Edwin Land of Polaroid in 1934 did some of the earliest work on electrophoretic devices. We are beginning to see versions of this technology being applied to electronic paper³⁴. A suspended particle device consists of 3-5 layers. The active layer has needle shaped dipole particles (<1 mm long) suspended in an organic fluid or gel. This layer is laminated or filled between two electrical conductors. In the off condition the particles are random and light absorbing. When the electric field is applied, the particles align with the field, causing transmission to increase. Typical transmission ranges are 20-60%, 10-50%, 0.1-10%, with switching speeds of 100-200 ms. The voltage required for the device depends on thickness and ranges from 0-20 to above 150 V a. c.

Research Frontiers Inc. and its licensees are responsible for the commercial development of SPDs for goggles, eyeglasses and windows. Recent activities have been directed at encapsulating the particles in a polymer sheet³⁵. Several companies have licenses with RFI for the development of specific products. Two commercial groups working with RFI are Hankuk Glass (Korea) and Materials Research Corp. (San Diego, CA). Their product focus is architectural and automotive glazings. Hankuk glass has been very innovative in developing prototype products for flexible plastic and large panels of about 1m²³⁶. The transmission properties of a Hankuk panel are shown in Fig. 6.1. It appears that electrophoretic technology is developing rapidly and is becoming viable for a variety of applications. Toyota Labs (Japan) is developing a new type of dispersed particle window. By modification of the particles, several colors (green, blue, red, and purple) can be achieved³⁷. These developments are beginning to make this technology that has been developed over 20 years much more viable for large-area glazing application. New submicron dipole suspensions based on SiO₂ coated TiO_xN_y have been developed by Nippon Sheet Glass³⁸. With this particle up to 50% change in solar transmittance has been shown.

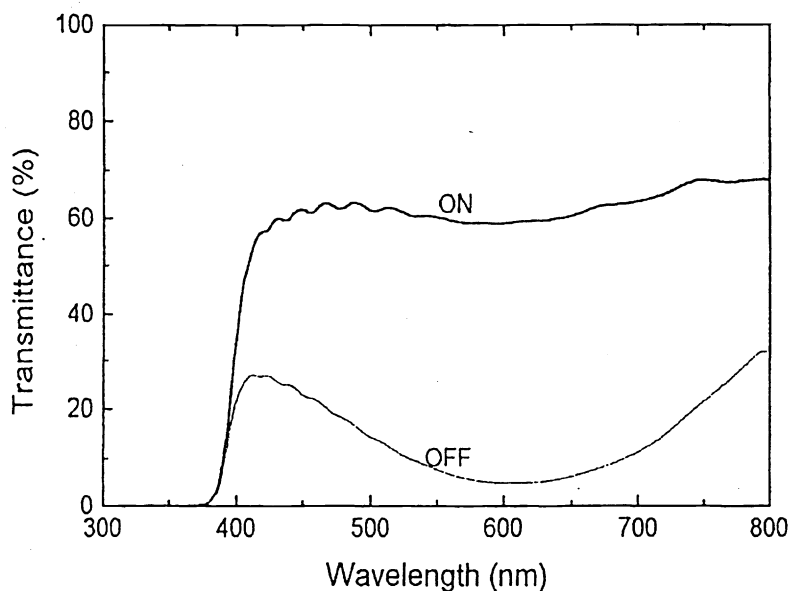


Fig. 6.1 Transmittance of an SPD Panel made by Hankuk Glass. Both the on and off conditions are shown. Tv=15% off and 60% switched on at 100 V a.c. (Source: Hankuk Glass, Korea)

7.CONCLUSIONS

Large-area switching films have unique properties for applications for large-area display, electronic paper and glazing. In this study selected technologies were covered including electrochromics, dispersed particles, electrophoretic displays, encapsulated liquid crystals, ferroelectric liquid crystals and the "Gyricon" bichromal ball display. Electrochromics are favored for many applications because when they switch they remain specular, and non-scattering. This means they can be used for a variety of view or see-through applications. They can be easily powered because of their low voltage. Electrochromics have been commercialized for automotive mirrors. Fairly large windows have been shown and installed in buildings by Pilkington/Flabeg, and Asahi Glass. Other companies are working on the introduction of glazing products for automotive sunroof applications. Saint-Gobain is working with Airbus to produce electrochromic passenger windows. Both Phillips, Saint-Gobain and Donnelly have made a prototype electrochromic contrast enhancement filters for the face of a CRT. Electrochromics for windows are still being evaluated for long-term durability. Production cost and simplification are major issues for large-area electrochromics. Donnelly, NREL and EPFL in Switzerland are developing "self-powering" photoelectrochromic windows. In one state, PDLCs are scattering and can not be used for view windows. PDLC's are sold on their unique privacy properties for security windows, utility vehicle side windows, and office privacy dividers. PDLC can be made in a flexible sheet form, but are limited to mainly interior applications because of UV stability problems. The chief maker of PDLC panels is Nippon Sheet Glass. A new panel based on cholesteric liquid crystals has been shown by CLCEO. This panel has lower scatter compared to regular PDLCs. Suspended particle technology (SPD) is similar to the PDLC's in that they are more absorbing or scattering in the off-state compared to the on-state. SPDs have an advantage of having much lower scattering in the off-state compared to PDLC's so they can be used for goggles and glasses. Also, SPD's can be made to have a neutral color. SPDs can be made into a flexible sheet form so they can be used in a variety of applications. Research Frontiers Inc., Hankuk Glass and Materials Research Corp. are working to produce a flexible product in the near-term. Hankuk has shown prototypes of displays, sunroofs and architectural windows. The cost is expected to be considerably lower than PDLCs.

Several unique technologies are being developed for the display and electronic paper markets. A future display market is electronic paper, where information could be stored and displayed on a flexible substrate. Electrophoretic displays are being developed by E-ink for that application. They are bistable and exhibit a memory effect. Another electrophoretic emulsion development for electronic paper is being developed at NOK Corp. Zikon is developing a reverse emulsion nanosize electrophoretic display for electronic paper. Xerox PARC labs (Palo Alto, CA) is developing a flexible bichromal ball display sheet called the "Gyricon". Certain thin film display technology is directed at improving the quality of CRTs. A flexible display sheet using ferroelectrics is under development for CRTs by Idemitsu Kosan in Japan.

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