Switchable Glazing: Science and Technology of Smart Windows

Carl M. Lampert

Lawrence Berkeley Laboratory, Energy and Environment Division, University of California,
1 Cyclotron Rd., MS 62-203, Berkeley, CA 94720 USA,
(510) 486-6093, Fax: (510) 486-7339, email: CMLampert@lbl.gov.

Keywords: Electrochromic coating, Tungsten oxide, Nickel oxide, Glass, Polyethylene teraphthalate (PET)

ABSTRACT

Electrically activated switchable glazing and their use as smart windows and other large-area applications are discussed. Electrochromic devices are compared to dispersed liquid crystals and dispersed particle glazing systems. A selection of device structures and performance characteristics are compared. A discussion of transparent conductors is presented. The characteristics of prototype and commercial devices from commercial and university labs in Japan, Europe, Australia, and USA are covered. A discussion of the future of this technology is made including areas of necessary development for the realization of devices in excess of 1 m^2.

INTRODUCTION

Switchable technology or "smart windows" has a very natural place in future glazings, it probably is one of the most exciting topics in glazings and has the potential to change the view of glazing from a fixed element to a dynamic dimable glazing. In the last few years there has been growing interest in this technology and is expected to continue well into the next century for a variety of products. There are various physical optical techniques that can be used for the regulation of incident daylight, solar energy, and glare. Applications include glazings in buildings, vehicles, aircraft, spacecraft and ships. The function of a smart window is to control the flow of light and heat into and out of a glazing, according to occupant comfort. Optical switching devices can also regulate lighting and heating levels for energy load mangement. Energy modeling of electrochromic windows has shown that electrochromic windows can provide significant energy performance improvement conpared to conventional double glazed windows [1-4].

The use of flat glass is very large, the world production of flat glass is about 2 billion m^2 per year. In the U.S. alone, glass production is about 0.5 billion m^2 per year (1994), with the largest portion going to building and automotive glazing. Electrochromic products currently in the marketplace are automobile and truck mirrors, and sunglasses. The mirrors are designed to automatically regulate glare in response to incident light levels. The mirrors are produced by Gentex (Zeeland, MI) and Donnelly (Holland, MI) with partnership with OCLI (Santa Rosa, CA). Electrochromic sunglasses were introduced as a product by Nikon (Tokyo, Japan) in 1994. Prototype glazing panels are being tested for automobile sun-roofs and visors. Near-future applications include automobile side and rear windows, architectural glazing, aircraft windows and perscription eyeglasses. Switchable glazing can also be used for large-area information displays in applications where high switching speed is not required, such as for airport display boards. One of the most significant issues is the cost of these devices and the trade-offs between cost and benefit, and cost and lifetime. The cost of an electrochromic window cost has been estimated to range from 100-1000 US$/$m^2$. Some companies have set cost goals of 100-250 US$/$m^2$. Both dispersed liquid crystal and dispersed particle windows fall within this range too. Current electrochromic development is aimed at long-life devices with durability similar to regular coated windows. A different approach is the development of low-cost limited-life switching devices. This is an area of important parallel development activity. There is growing investment by larger companies and national governments in promoting this technology. Specifically some projects are in Japan, Europe, Australia and in the U.S.

The basic property of an electrically activated smart window material is that it exhibits a large change in optical properties upon a change in either electrical field or injected or ejected charge.The change in optical properties can be in the form of absorptance, reflectance or scatttering. This optical change results in a transformation from a highly transmitting state to a partly reflecting or absorbing state. This change can be either totally or partly over the visible and solar spectrum. Typically it is over some portion of the spectra. The electrically activated devices has the advantage of user or automatic control. There are other switchable materials, such as photochromic (glass and plastic) and thermochromic devices (polymer gels and metal oxides) that are not covered in this study but are covered in other work [5.6].

Thin and thick film fabrication is relevant to all of the switchable glazing technologies. Several large-area deposition technologies are necessary, such as planar reactive d.c. magnetron sputtering, different types of chemical vapor deposition, and sol-gel coating. Since electrochromic and conductor layers are fairly thick, of the order of 200-400 nm

Society of Vacuum Coaters 505/856-7188
[illegible] Annual Technical Conference Proceedings (1995)

thick for each active layer, high rate is important. These coatings are about 10 times thicker then the typical interference coating. High quality is important since electrically activated coatings don't tolerate pin holes. Fortunately, high rate sputtering is advancing rapidly. The advent of rotating cathodes, with superior materials utilization, leads to a drop in manufacturing cost. Chemical vapor deposition (spray pyrolysis) makes it possible to coat directly on the glass ribbon of the float line in the manufacturing plant. This process can give a very inexpensive coating. Currently, it is used to deposit transparent conductors (low-e coatings). Also, plasma enhanced chemical vapor deposition lends itself to very high deposition rates for certain compounds. Advances in fabrication technology will help reduce the cost of switchable windows. With these technologies there are still significant challenges for the process designer to adapt large-scale deposition technologies to the fabrication of square meter size multilayer switchable glazing with high optical quality.

A significant issue for all electrically activated devices is the quality and cost of transparent conductors. Examples are $In_2O_3:Sn$ (also known as ITO) and $SnO_2:F$. The transparent conductors are a dominate cost of the switchable glazing, and necessary for all device types [7,8]. There is considerable development on the production of low-resistivity ITO or ZnO transparent conductors deposited onto plastic substrates at low temperatures [9]. Fairly new coated glass products for Low-e surfaces, e.g., Tech 15 Glass (LOF Glass, Toledo, OH), K-Glass (Pilkington Glass, UK), can be used for electrochromic windows. The cost in quantity is about $\$15/m^2$ which is considerable less than electronic grade ITO/glass. It consists of a low resistivity coating of doped tin oxide produced by thermal pyrolysis directly on the glass float line. Pilkington uses this material for their prototype electrochromic windows.

ELECTROCHROMICS

Electrochromic windows are a very popular area of all switching technology. Over the last ten years about 200 U.S. and international patents have been granted per year. The major advantages of electrochromic materials are: (1) they only require power during switching; (2) require a small voltage to switch (1-5 V); (3) are specular under all conditions; (4) have contineous diming; (5) many designs have a long term memory (12-48 h). Typical electrochromics have upper visible transmission of T_v=0.65-0.50 and fully colored transmittance T_v=0.25-0.10. Lower levels of transmittance have been noted below 1% but only in very specialized devices. The shading coeficient change is about SC=0.67-0.60 for the bleached condition, and SC=0.30-0.18 for the fully colored condition.

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 is:

WO_3 (transparent) + yM^+ + ye^- <-> M_yWO_3 (blue) where $M = Li^+, H^+, Na^+, Ag^+$, etc.

A typical anodic reaction is:

$Ni(OH)_2$ (transparent) <-> NiOOH (bronze) + H^+ + e^-

The inorganic mateiials that have gained the most research interest are various forms of WO_3, NiO, MoO_3, and IrO_x. These compounds, among other transition metal oxides, are the subject of several technology reviews [10-15]. An electrochromic device must use an ion-containing material (electrolyte) in close proximity to the electrochromic layer as well as transparent layers for setting up a distributed electric field. Devices are designed in such a way that they shuttle ions back and forth into the electrochromic layer with applied potential. An electrochromic window can be fabricated from five (or less) layers consisting of two transparent conductors, electrolyte or ion conductor, counter electrode, and electrochromic layer. Depending on the components used in devices, some of the layers can be combined serving dual functions. Some devices use even more layers depending on design. Ion conductors and solid electrolytes also require further development for this technology. The most promising ion conductors are certain immobile solvent polymer systems, ionic glasses and open channel metal oxide structures such as Perovskites. Typical electrochromic device structures developed by industry and university groups are given in Table I.

Organic electrochromics are based on the viologens, anthraquinones, diphthalocyanines, and tetrathiafulvalenes. 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 recently more stable organic systems have been developed. Both the Gentex Co. and Donnelly Co. have commercially developed organic electrochromic materials for automotive mirrors. The truck mirror commercialized by Donnelly is based on $H_yWO_x/Ta_2O_5/NiO_x$. Mirrors are the most commercially developed electrochromic product to date. Gentex has produced over 1 million mirrors over the last few years. The next area of automotive electrochromic development is for sunroofs and visors. St. Gobain Glass (Paris, France) is developing electrochromic sunroofs for French automakers. Donnelly has developed a sunroof for the Ford Motor Co. This is an ideal large-area entry market

Table I Electrochromic Device Structures

Electrochromic	IC/Electrolyte	Ion Storage	Maker	Application
Proton Systems				
Viologen	PMMA+Organic	none	Gentex	Auto Mirrors
a-H_xWO_3	Ta_2O_5	NiO	Donnelly	Auto/Truck Mirrors
a-H_xWO_3	Ta_2O_5	$Ir_xSn_yO_2$:F	Nikon	Auto Mirrors
a-H_xWO_3	SiO_2/metal	WO_3	Schott	Auto Mirrors
a-H_xWO_3	Polymer	Polyaniline	Toyota	Auto Glazing
a-H_xWO_3	Polymer	Polyaniline	Dornier	Aircraft Glazing
a-H_xWO_3	Poly-AMPS	Cu Grid	PPG	Aircraft Glazing
a-H_xWO_3	Ta_2O_5	a-IrO_2	EIC	Spacecraft Glazing
a-H_xWO_3	a-PEO copolymer	Redox Polymer	LBL	Building Glazing
Lithium Systems				
a-Li_xWO_3	$LiClO_4$+ PC	Prussian Blue	Central Glass	Glazing
a-Li_xWO_3	metal oxide	NiO	Asahi	Building Glazing
a-Li_xWO_3	Li-PEO	CeO_x	St. Gobain	Auto Glazing
a-Li_xWO_3	Li-B-SiO Glass	Li_xCrO_4	EIC	Building Glazing
a-Li_xWO_3	PPG-$LiClO_4$-MMA	$Li_yV_2O_5$	Chalmers U.	Building Glazing
a-Li_xWO_3	Li-PEO	NiO	St. Gobain	Auto Glazing
c-Li_xWO_3	$LiNbO_3$	Li_xCoO_2	Tufts	Building Glazing
a-Li_xWO_3	modified a-PEO	Ion Storage Polymer	LBL	Building Glazing
a-Li_zWO_3	Li-a-PEO	Li_xNi_yO	LBL	Building Glazing
a-Li_xWO_3	Li-polymer	LiV_2O_5	NREL	Building Glazing

because of the smaller size and shorter lifetime required of the window compared to building glazings. Although the upper temperature limits are higher (90-120 C), according to standard desert static soak temperatures for cars.

Several research groups are investigating electrochromic materials and devices for building windows [16]. Pilkington PLC (UK) has a multiyear project with the Commission of the European Communities (CEC) on electrochromic glazing under the JOULE II programme. This project involves several organizations, including Flachglas AG (Germany) and Danionics (Denmark). Flachglas has recently shown a prototype switchable glazing greater than $1m^2$. Under this project laminated glazings have been developed based on Li_xWO_3/Li-polymer/NiO,TiO_2. Asahi Glass (Yokohama, Japan) has been steadily developing prototype electrochromic windows (0.6 x 0.8 m) based on Li_xWO_3/metal oxide/NiO_x for testing and evaluation. Part of Asahi's work has been funded by the Japanese govenment-Sunshine project under MITI. About two hundred (0.4 x 0.4 m size) prototype electrochromic windows have been installed in the Seto Bridge Museum (Kojima, Okayama-Pref., Japan). In Australia, a govenment grant has funded the Univ. of Technology (Sydney), Glassform (Victoria) and Silicon Technologies (Queensland) to develop sol-gel deposited electrochromic devices.[nnn] These devices are based on laminated structures using WO_3. There are efforts at Sony and Sharp in Japan to develop a multicolor flat panel display using organic electrochromics. New techniques for the electrodeposition of polymer electrochromics from solution have been reported by Toyota of Japan [17]. Uniform films of polythiophene, polyaniline and polypyrrole have been deposited by a oxidation polymerization technique. Polyaniline is one of the favored organic polymer electrochromics [18,19].

Under the U.S. DOE (Dept. of Energy) Electrochromics Initative both Donnelly, and OCLI (Santa Rosa, CA) in partnership with EIC Labs. have contracts to develop large-area electrochrochromic glazing. Also, under the U.S. Dept. of Commerce, NIST grant, the partnership of 3M (St. Paul, MN) and SAGE Corp. (Piscataway, NJ) was awarded a sizable grant to develop electrochromics on plastics. At LBL we serve as the lead lab for the U.S. Dept. of Energy Program. We have developed devices and components for mainly laminated devices based on WO_3, NiO, NbO_x and Li_xNiO.[20-22] Another development at LBL has been on polymer ion storage layers for lithium and proton storage [23]. The development of testing and evaluation procedures has been done through IEA (The International Energy Agency) [24] and at NREL (Golden, Co) [25]. A different approach is the development of low-cost switching devices based on plastic substrates. Southwall Technologies (Palo Alto, CA) has produced electrochromic electrodes of tungsten oxide/ITO on polyester. Over the next few years we expect to see sizable development in this field.

Certain forms of crystalline 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. Crystalline WO_3 is being studied by several investigators [26-28]. Peak near-infrared switching levels from 20% to greater than 75% has been achieved with crystalline tungsten oxide. The reflectance properties of tungsten oxide films produced so far seem to lie far from the theoretically limiting behavior.

LIQUID CRYSTAL GLAZING

Liquid crystal based systems offer another approach to electrically switchable glazings. The basic classes of liquid crystals are the twisted nematic, guest-host, surface stabilized ferroelectric, and polymer dispersed liquid crystals. So far polymer dispersed liquid crystals are the only ones used for commecial glazings. The guest-host liquid crystals are another choice for large-area glazing. The mechanism of optical switching in liquid crystals is to change the orientation of liquid crystal molecules interspersed between two conductive electrodes with an applied electric field. The orientation of the liquid crystals change with the field strength that alters the overall optical properties of the window. Open circuit memory is not possible with liquid crystals.

The polymer dispersed liquid crystal (PDLC) or encapsulated liquid crystal (NCAP-nematic curvilinear aligned phase) consists of nematic liquid crystals distributed in microcavities [29,30]. This cavity structure can give modulated light scattering known as the Christiansen effect. PDLC and NCAP materials have very similar characteristics but are defined in the patent literature differently according to preparation procedure. NCAP films are formed from an emulsion and PDLC films are formed from an isotropic solution which phase separates during curing. 3M (St. Paul, MN) with General Motors Research Labs (Warren, MI), and Kent State University (Kent, OH) have developed PDLC technology for automotive glazing [31]. Their products have been available since 1994.

Devices based on NCAP technologies were produced by Raychem (formerly by Taliq) (Sunnyvale, CA) as optical shutter materials for glazings and information displays (known as Varilite and Vision Panel). Nippon Sheet Glass (Itami, Japan) markets this product in Japan as the "Umu" Device. Large-area devices have been fabricated in 1 m x 2.5 m sheets. With NCAP the liquid crystals are encapsulated within an index matched polymer matrix. The composite polymer is fabricated between two sheets of ITO coated polyester (PET) that serves as electrodes. The device can also be fabricated between one sheet of glass and one sheet of plastic or two sheets of conductive glass. 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. Since the off-state in these devices is diffusely transmitting, the device has application for privacy and security. The dyed film shows considerable control over visible transmittance compared to an undyed film. When an electric field is applied, the liquid crystal droplets align with the field and the device becomes transparent [47]. Typically these devices operate between 60-100 V a.c., (potentially lower in the future) at less than 20 W/m^2 but require continuous power to be clear. The typical integrated hemispherical visible transmission values for a 3M-PDLC device are $T_{visible(off-on)}$= 0.49-0.80. The shading coefficient changes by SC=0.63-0.79 [32]

Pleochroic dyes can be added to darken the device in the off-state [33]. In general, compared to electrochromics, the power consumption is higher for liquid crystals because of the need for continuous power in the activated state. Dispersed liquid crystal devices have a very good future but are restricted by three characteristics: the unpowered state is diffuse, haze remains in the activated (transparent) state and UV stability is poor. For widespread use of NCAP and PDLC devices the cost will have to decline.

The guest-host liquid crystal uses dichroic dye molecules mixed with liquid crystals [34]. The guest-host type is being developed mainly for display applications, but has been considered for optical shutter applications. The cholesteric-nematic phase change guest-host type of liquid crystal has a good chance of being used for large-area optical switching. This type of liquid crystal switches by a field induced phase change. It exhibits a continuous gray scale operating from 20-120 a.c.V with a contrast ratio of 5:1 and uses no polarizers. The upper temperature stability is 100 C. Guest-host liquid crystal devices are being developed by Asulab in Switzerland for eyeglasses and car mirrors. A car mirror based on this technology can switch visible reflectance, R_v= 0.48 to 0.12 with switching times of 15 and 360 ms for on and off respectively [35]. This has been possible with the development of improved electrochemical and UV stability of the liquid crystals.,

DISPERSED PARTICLE GLAZING

The development of suspended particle or electrophoretic devices has spanned many years. Some of the earliest work was done by Edwin Land of Polaroid in 1934. The development of suspended particle devices has been slowed by a number of technological problems including long-term stability, cyclic durability, particle settling and agglomeration, and gap spacing control for larger glazing. In recent years, many of these problems have been reduced or controlled [36-38]. Commercial development of these devices for goggles, eye glasses and windows is being done chiefly by Research Frontiers, Inc. (RFI) (Plainview, NY) and it's licensees. Recent developments have been to encapsulate the particles in a polymer sheet. A group of 50 RFI windows (0.46 x 0.46 m) have been installed as a demonstration in the Japan Steel Works Building (Fuchan, Japan). Gloverbel, S.A. (Brussells, Belgium) is working to develop mirrors and windows for cars. Commercial groups working on this technology are are Hankuk Glass Industries (Korea), Northern Engraving for automotive displays and Litton Systems Canada for visors [39]. Another version of this device is being developed by Nippon Sheet Glass in Japan. [40]

A suspended particle device consists of 3-5 layers. The active layer has needle shaped particles of polyiodides (dihyrocinchonidine bisulfite polyiodide) or paraphathite ($1\mu m$ long) suspended in an organic fluid or gel. This layer is laminated or filled between two parallel 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.

CONCLUSIONS

Switchable glazings will ultimately lead to better building and vehicle glazing, and display technology. In a number of uses, it will add to the comfort and well-being of its users. Smart windows will help lower of the demands on precious non-renewable fuels for lighting and cooling. The prospects are great with many major companies involved in the development of switchable glazings worldwide. Also, the scientific and technological challenges of the development of these complex glazings are large, creating a specialized area of research. In this study, electrochromic devices were compared to dispersed liquid crystals, and suspended particle devices. Of the three major types of electronically activated glazings all three have their particular applications. There are several promising electrochromic devices for window glazings. For the electrochromic layer, tungsten trioxide, nickel oxide, iridium oxide and viologens are the most developed for devices. In the future, the developments will lie in the designing of the appropriate ionic and electronic materials to make devices durable over many years and cycles. Dispersed liquid crystal devices have a very good future but are restricted by three characteristics: (1) the unpowered state is diffuse, (2) haze remains in the activated (transparent) state, and (3) UV stability is poor. The development in dispersed particle devices is moving forward with a number of commercial activities and demonstration windows. These devices are colored in the off-state and become transparent in the on-state. With further improvements in processing and the development of a sheet form of this material, these devices could see widespread use. Durability is of importance for all smart window devices, and an expected lifetime of at least 5-15 years must be assured. The great challenge is to make these technologies, through advanced deposition and fabrication techniques, economical in the 100-250 $\$/m^2$ price range. But still, optical switching glazings are emerging in a few products. It is expected their application will greatly redefine our concept of the window.

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ACKNOWLEDGMENTS

The author wishes to thank The SVC organizing committee especially Dale Martin and notably Clark Bright, Southwall Corp., for inviting this paper. I wish to thank the cited authors and glass and window company representatives who provided me with advice and technical information for this study.

This review was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Building Technologies, Building Systems and Materials Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.