Electrochromics for energy efficiency and indoor comfort*

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Abstract: Electrochromic (EC) oxide films with nanocrystalline and nanoporous structure constitute the basis of a number of devices for modulating the throughput of radiant energy. One example is in architectural windows capable of providing energy efficiency and indoor comfort simultaneously. This paper introduces EC technology with particular attention to applications to the built environment. The nanostructural features of the EC films are emphasized, and it is pointed out that reactive magnetron sputtering can be used for large area manufacturing. Electrochromics is discussed from the viewpoints of materials, device technology, and applications.

Keywords: electrochromics; thin films; tungsten oxide; nickel oxide; energy efficiency; buildings.

INTRODUCTION

Ambient radiation is characterized by three general features: spectral selectivity, angular dependence, and temporal variability [1–3]. Regarding spectral selectivity, solar radiation onto the atmospheric envelope comes in the $0.2 < \lambda < 3$ μm wavelength range, while thermal radiation at normal temperatures lies at $3 < \lambda < 50$ μm, and visible light as well as light for photosynthesis is at $0.4 < \lambda < 0.7$ μm. The atmosphere is transparent in the main parts of the $0.3 < \lambda < 3$ and $8 < \lambda < 13$ μm intervals if the humidity and cloudiness are low. These facts of nature lead to many possibilities to develop "solar energy materials" with optimized absorptance A, transmittance T, reflectance R, and emittance E [1,2]. For example, efficient solar heating requires, ideally, $A = 1$ for $0.3 < \lambda < 3$ μm and $E = 0$ for $3 < \lambda < 50$ μm, and efficient cooling via radiation to the clear sky demands $R = 1$ for $0.3 < \lambda < 8$ and $13 < \lambda < 50$ μm, while $E = 1$ should prevail for $8 < \lambda < 13$ μm. Concerning architectural windows, $T = 1$ for $0.3 < \lambda < 3$ μm and $E = 0$ for $3 < \lambda < 50$ μm give maximum solar heating jointly with thermal insulation for double glazings, whereas $T = 1$ for $0.4 < \lambda < 0.7$ μm and $R = 1$ for $0.7 < \lambda < 50$ μm give full day-lighting at minimum solar heating. Optimized angular properties can boost the performance of spectrally selective surfaces. These and other aspects of solar energy materials have been reviewed recently [1–3].

It is also possible to design for temporal variability by exploiting "chromogenics" [4], which includes photochromic materials (typically darkening under irradiation in the $0.3 < \lambda < 0.4$ μm range), thermochromic films (typically increasing R at $0.7 < \lambda < 3$ μm as the temperature exceeds a certain "critical" value in the vicinity of room temperature), "gasochromic" films (darkening/bleaching under

exposure to reducing/oxidizing gases), and electrochromic (EC) multilayer structures [5–8] (changing their absorption at $0.3 < \lambda < 3$ µm under charge insertion/extraction).

The purpose of this paper is to briefly outline EC device technology—with a view to the nanostructure of its optically functional layers—and discuss possibilities to revolutionize building technology by allowing rigid or flexible building envelopes that are able to adjust their throughput of visible light and solar heating in order to combine energy efficiency and indoor comfort. In particular, this technology is capable of alleviating air conditioning loads that are currently much in focus owing to their rapid increase [9,10]. The growth, concerning the electrical peak load, lay at an unsustainable value of ~17 % per year in the EU15 between 1995 and 2003 [11]. Another example of many can be found in a recent study from Kuwait reporting that more than 75 % of the electricity is consumed by air conditioning at peak load, and that this load has increased by a factor ~2.5 from 1991 to 2001 [12]. The present paper is partly based on an earlier conference report [13].

FUNDAMENTALS OF ELECTROCHROMICS-BASED BUILDING ENVELOPES

Energy efficiency

It is common for people to spend ~90 % of their time inside buildings and vehicles, and clearly the quality of the indoor environment is of great importance. Increasing amounts of energy are used to maintain this environment at a level that is both comfortable and healthy. Looking at the EU15, some 40 % of the energy supply is currently used for heating, cooling, ventilation, and lighting of buildings, which corresponds to ~4 % of the gross national product [14,15].

The energy efficiency accomplished by the use of EC-based building envelopes has been difficult to assess, partly since the primary function of a window or glass façade—to give unmitigated visual contact between indoors and outdoors—has not always been sufficiently well understood. The most detailed investigation so far of this inherently difficult and multi-faceted issue is found in recent work for the California Energy Commission [16,17]. It states that, using today's best fenestration technology as a baseline, EC windows showed annual peak cooling load reductions from control of solar heat gains of 19–26 %, and lighting energy use savings of 48–67 % when controlled for visual comfort. Furthermore, subjects strongly preferred EC windows over reference windows, with preferences related to perceived reduction in glare, reflection on computer monitors, and window luminance. These numbers on the energy savings—although huge—are likely to be underestimated since they account neither for novel transparency control strategies based on physical presence [18] nor for novel day-lighting strategies based on "light balancing" [19].

Alternative, though less detailed, estimations of the energy efficiency of EC-based building envelopes have been investigated in recent simulations [20,21]. The energy for space cooling, on an annual basis, could be reduced by as much as 40–50 % when the EC-based technology was implemented. An interesting result was that air conditioning might be avoided when "smart windows" are used, thus indicating that the marginal cost for "smart windows" can be more than balanced by the elimination of an air conditioning system. Results of even simpler "back-of-an-envelope-analyses" for the energy efficiency [18] emphasize an analogy between EC-based windows and today's best solar cells, having the same size and orientation: the solar cells can *generate* a certain amount of energy, which is similar to the energy *savings* the "smart windows" can provide.

Device design and materials

An EC device resembles a thin-film electrical battery, as evident from Fig. 1. There are five superimposed layers on a transparent substrate—normally of glass or flexible polyester foil—or positioned between two such substrates in a laminate configuration [5,8]. The outermost layers are transparent electrical conductors, typically of In_2O_3:Sn (i.e., indium tin oxide, denoted ITO) [3,22]. One of these layers
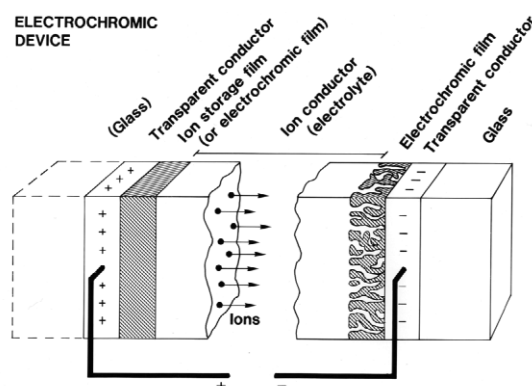
Fig. 1 EC device design and illustration of transport of positive ions under the action of an electric field. From [5].

is coated with an EC film, and the other is coated with an ion storage film, with or without EC properties. Both of these films must consist of nanomaterials with well-developed nanoporosities (as in battery electrodes). A transparent ion conductor (i.e., electrolyte) is at the middle of the device and joins the EC and ion storage films. Voltage application during a specified time between the transparent electrodes leads to ion exchange between the EC and ion storage films, and the overall transparency is then changed. A voltage with opposite polarity—or, with suitable material combinations, short circuiting—brings back the original properties of the device. The optical modulation requires a DC voltage of 1–2 V. The ion insertion into the EC film(s) is balanced by electron inflow from the transparent electrode(s); these electrons can produce intervalency transitions, which is the basic reason for the optical absorption [5,8].

Concerning materials in EC devices, the ITO can be replaced by $ZnO:Al$, $SnO_2:F$, or similar oxides (or, possibly, carbon nanotubes) [3,23,24]. The global availability of indium has been much debated recently, following steep price hikes. The situation is now becoming clear, and it appears that the demand is ample and larger than that of silver [25,26].

The EC film is based on WO_3 in almost all devices for architectural applications, while there are many options for the counterelectrode [5–8]. Among the latter, films based on IrO_2 and NiO have received much interest recently. IrO_2-based alternatives are inherently expensive, but good EC properties are maintained after dilution with cheaper Ta_2O_5 [27]. NiO-based films combine moderate cost with excellent optical properties, especially when the NiO is mixed with another oxide characterized by a wide bandgap such as MgO or Al_2O_3 [8,28]. EC devices can use many different electrolytes such as hydrous oxides exhibiting proton conduction or polymers with ion conduction by added salts.

EC devices have been discussed a long time, originally for non-emissive displays [29] and subsequently for rear-view mirrors and windows [30]. Some applications have emerged recently [31], but, broadly speaking, the progress of EC-based technologies has been slow, which may be associated with the necessity of successfully combining several nonconventional technologies as follows: (i) the ITO must have excellent electrical conductivity together with very low optical absorption, which is challenging particularly for films on temperature-sensitive substrates such as polymeric ones [3]; (ii) the EC and counterelectrode films must exhibit uniform nanoporosity over large areas; (iii) viewing the EC device as a "thin-film battery" makes it evident that charge insertion/extraction and charge balancing must be accomplished by properly controllable and industrially viable techniques, such as facile gas treatments [32]; (iv) the electrolyte must combine good ion conductivity with adhesiveness and high transparency for luminous and ultraviolet radiation; and (v) long-term cycling durability demands adequate strategies for voltage and current control during coloration/bleaching, in analogy with the situation for charging/discharging of batteries. These challenges can be successfully met, however, so that EC-based technology finally may emerge as suitable for large-area, large-scale applications [8,31].

Film porosity is necessary on the nanoscale, as stressed in (ii) above. Most thin-film deposition technologies may be capable of achieving this, though with more or less difficulty. Regarding sputtering, the nanostructure can be inferred from the well-known "Thornton diagram" [33] illustrated in Fig. 2. Thin films most often are prepared under conditions corresponding to the "transition zone" denoted T. Those films are compact, and in a metallic layer it is possible to minimize grain-boundary scattering of the conduction electrons, which is important for ITO transparent conductors [34]. Nanocrystallinity and -porosity are found at higher pressure in the sputter plasma, such as in "zone 1". Here it is possible to have ion conduction in the inter-columnar spaces, as required in EC films and in solid-state ionic films in general.
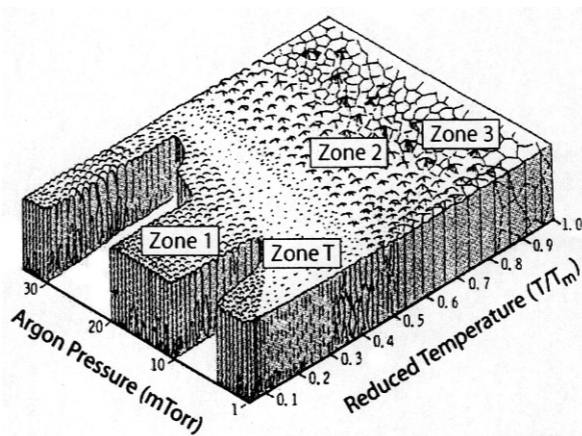

Fig. 2 "Thornton diagram" illustrating nanostructures of thin films prepared by sputtering at different argon pressure and substrate temperature. The melting point of the material is denoted T_m. From [33].

PROPERTIES AND APPLICATIONS OF ELECTROCHROMIC FOILS

Thin-film deposition

Reactive DC magnetron sputtering from targets of tungsten and nickel was used to make the devices discussed below [8,35]. An addition of vanadium, magnesium, or aluminum to the nickel was used in many experiments; this made the targets nonmagnetic and hence convenient for magnetron sputtering. Deposition took place in $Ar/O_2/H_2$ with optimized mixing ratio. The target–substrate distance typically was 20 and 25 cm, and the total gas pressure was in the 30–40 mTorr range (cf. Fig. 2).

From a manufacturing perspective, it is preferable to be able to deposit the films under conditions that make them ready for device assembly, or to have convenient processes to prepare the as-deposited films for this. In the case of tungsten oxide, deposition in $Ar/O_2/H_2 = 4/2/13$ is suitable and leads to bluish-colored films. A key issue with regard to optimizing the gas mixture is to incorporate hydrogen into the film without creating oxygen deficiency. The nickel-oxide-based film was post-treated in ozone [32].

Optical and electrical properties

Figure 3 shows typical data for a 5×5 cm^2 flexible EC device incorporating two laminated 175-μm-thick poly(ethylene terephthalate) (PET) foils, a WO_3 film, a NiO film modified by addition of a wide-bandgap oxide such as MgO or Al_2O_3, poly(methyl methacrylate) (PMMA)-containing electrolyte, and ITO layers [36]. The mid-luminous transmittance (T_{550}, where the subscript denotes wavelength in nanometers) rapidly reaches ~68 % upon bleaching and decreases to ~36 % during a coloration period
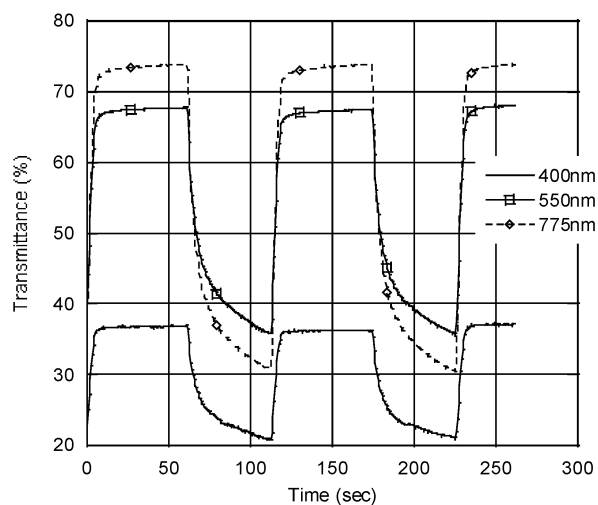
Fig. 3 Time-dependent optical transmittance during coloration and bleaching of an EC device, recorded at three wavelengths. From [36].

of 50 s. Much lower values can be obtained with extended coloration times. The modulation is smaller, but still significant, for blue light (T_{400}) whereas it is larger for red light (T_{775}). The substantial transmittance at the UV end of the spectrum is associated with the oxide added to the NiO. Durability was observed for several tens of thousands of coloration/bleaching cycles. The open-circuit memory is excellent, and the optical properties can be maintained almost unchanged for many hours.

If the transmittance of the EC-based device needs to be enhanced, it is possible to apply antireflection coatings. Self-assembled silica nanoparticle films are of particular interest and makes it possible to reach a transmittance as large as 99.5 and 98.6 % at a mid-luminous wavelength for double-sided coating of glass and polyester foil, respectively [37,38]. Specifically, a flexible EC-based foil was immersed in a sol comprised of ~50-nm-diameter SiO_2 nanoparticles in ethanol, was withdrawn at a rate in the interval 2–2.5 mm/s, and was then subjected to mild heating for 10 min. The heat treatment improved the mechanical properties of the antireflection layers, which withstood a simple Scotch tape adhesion test. Figure 4 reports on a PET-foil-based EC device in its bleached state. The mid-luminous transmittance was enhanced by 6 percent units as a result of the antireflection treatment. It should be noted that this EC device had a vanadium-containing counterelectrode, which limited the transmittance to a significant degree so that the data are not directly comparable with those in Fig. 3.
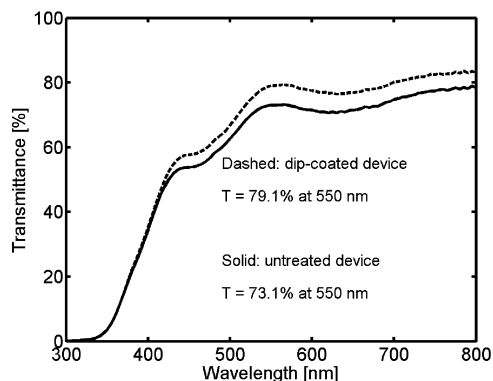

Fig. 4 Spectral transmittance T of an EC foil-type device with and without antireflection treatment. From [38].

Some applications

The EC foil can be used in a variety of applications related to buildings, vehicles, and consumer products. It can be suspended between transparent panes, employed as a conventional add-on "window film", or used as a lamination material between rigid glass or polymer panes. The latter application can give impact resistance, spall shielding, acoustic damping, etc., as added benefits. Figures 5, 6, and 8 show EC-based prototypes of windows in buildings, airplane windows, and visors for motorcycle helmets, respectively.

Fig. 5 "Smart window" prototype with four 30×30 cm^2 panes.

Fig. 6 EC-based airplane window prototypes.

Specifically, Fig. 5 illustrates a "smart window" with four 30×30 cm^2 EC foils mounted between glass panes. The upper two panels are fully colored while the lower two are fully bleached. Each of the panels can shift gradually and reversibly between the dark and transparent states in about a minute, i.e., during the time it takes for the eye to adapt to different lighting conditions. It should be noted that the window maintains its primary function—i.e., it provides unmitigated visual indoors/outdoors contact—irrespective of the state of coloration. This window cannot give privacy, albeit that might become feasible in constructions with superimposed foils [38]. Figure 6 illustrates two full-size airplane windows; the left-hand one is in a fully dark state and the right-hand one is in a fully transparent state. Figure 7 shows the mid-luminous transmittance of such a window [39]; the drive circuitry has a five-level con-
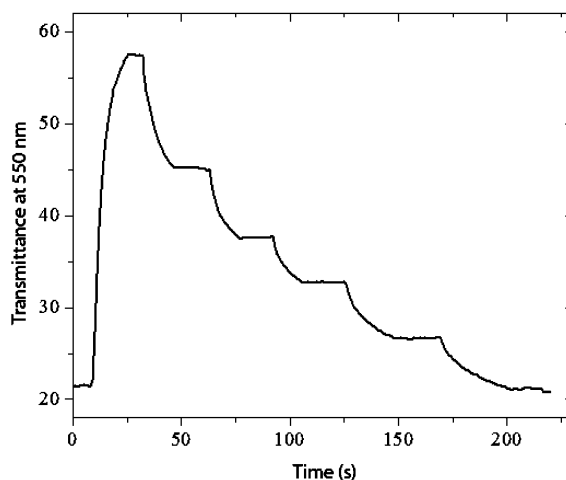
Fig. 7 Time-dependent mid-luminous transmittance of an airplane window prototype such as the one in Fig. 6. From [39].

troller allowing individual adjustments of the transmittance level. It is interesting to note that solar ir-radiation in itself has the effect of making the optical changes more rapid [38].

Potential applications in energy-efficient buildings and vehicles have provided the main impetus for research and development on EC for several years. However, there are numerous other applications as well, especially with regard to foil-type devices. They can be applied, for example, in ski goggles and in visors for motorcycle helmets. Such visors can be colored to a chosen degree in the day and bleached in the night, and driving and riding safety can be improved by having the visor bleach before entering tunnels or other dark spaces. Another aspect with a bearing on safety is that facial warming can be sig-nificantly limited by luminous absorption and infrared reflectance in the visor [40]. Figure 8 illustrates a visor in a motorcycle helmet.

Fig. 8 EC-based visor for motorcycle helmets.

PERSPECTIVES AND CONCLUSIONS

Electrochromism was discovered and made widely known many years ago [41]. The progress of EC de-vice technology has been slow, though, and a number of reasons for this were given above. Buildings with EC-based "smart windows" have been tried by the glass and coatings industry for decades, and more or less encouraging results have been reported. For one reason or another, these "smart windows" have not made it to the market. Today the scene seems to be changing, though, and EC-based roof win-

dows for buildings [42] and cars [43] are available on a limited commercial scale. These devices use glass as a base, implying that large substrates and expensive coating units must be used. However, cheap production of EC devices is necessary if the technology will be used elsewhere than in small niche markets. An alternative route, using web coating by technologies allowing roll-to-roll manufacturing, is now tested for EC prototypes [8]. This EC foil was discussed above.

Turning now toward the future, windows with tunable optical properties have long been something of a Holy Grail in "high-tech" architecture [44]. Electrically controlled EC-based "smart windows" are particularly interesting considering their potential to provide user-related operation. These windows make it possible to combine increased indoor comfort for the occupant of the building (less glare and thermal stress) with large energy efficiency (especially lowered air conditioning load in cooled spaces), as emphasized above, and hence there are strong incentives for their introduction. Durability, optical switching speed, and size constraints constitute an interrelated problem complex which is not fully solved today, but it seems to be increasingly accepted that a satisfactory solution can indeed be achieved. It should be noted that electrochromism is an enabling technology of relevance for a vast number of applications, so there may be multiple roads toward devices eventually used in buildings.

EC technology may be combined with light-guiding devices to direct daylight deeply into buildings. By equalizing the light level in a room, the eye—which tends to adjust to the brightest illumination—does not perceive deeper regions as disturbingly dark, and hence there is less need for artificial lighting. Similar notions, though employing a Venetian blind system, have been discussed recently [45,46]. Generally speaking, the EC technology leads to new opportunities for energy-efficient daylighting, which is generally regarded as superior to artificial lighting by giving better task performance, improved visual comfort, and positive mood effects, especially if glare problems are eliminated [47–49]. Particularly beneficial effects have been observed concerning increased sales in day-lit stores [50] and student performance in day-lit schools [51].

As a final note, we observe that history has provided convincing evidence that a new technology—such as EC-based foils—will lead to new applications in fields that may not be in any way self-evident today. This is sometimes referred to as "Kroemer's Lemma of New Technology" [52–54], which states:

> The principal applications of any sufficiently new and innovative technology always have been—and will continue to be—applications created by that technology.

One such possibility may be the merging of membrane architecture [55,56] and EC foil technology in order to allow light-weight buildings surrounded by large climate shells. Using ethylene tetra fluoroethylene (ETFE)—a well-known building material used for 25 years—the embodied energy in the climate shell can be an order of magnitude lower than for a corresponding glass-based construction [57].

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