Technology Advancements to Lower Costs of Electrochromic Window Glazings

Final Report

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Abstract

An Electrochromic (EC) Window is a solar control device that can electronically regulate the flow of sunlight and heat. In the case of the SageGlass® EC window, this property derives from a proprietary all-ceramic, intrinsically durable thin-film stack applied to an inner surface of a glass double-pane window. As solar irradiation and temperatures change, the window can be set to an appropriate level of tint to optimize the comfort and productivity of the occupants as well as to minimize building energy usage as a result of HVAC and lighting optimization.

The primary goal of this project is to replace certain batch processes for EC thin film deposition resulting in a complete in-line vacuum process that will reduce future capital and labor coats, while increasing throughput and yields. This will require key technology developments to replace the offline processes.

This project has enabled development of the next generation of electrochromic devices suitable for large-scale production. Specifically, the requirements to produce large area devices cost effectively require processes amenable to mass production, using a variety of different substrate materials, having minimal handling and capable of being run at high yield. The present SageGlass® production process consists of two vacuum steps separated by an atmospheric process. This means that the glass goes through several additional handling steps, including venting and pumping down to go from vacuum to atmosphere and back, which can only serve to introduce additional defects associated with such processes. The aim of this project therefore was to develop a process which would eliminate the need for the atmospheric process.

The overall project was divided into several logical tasks which would result in a process ready to be implemented in the present SAGE facility. Tasks 2 and 3 were devoted to development and the optimization of a new thin film material process. These tasks are more complicated than would be expected, as it has been determined in the past that there are a number of interactions between the new material and the layers beneath, which have an important effect on the behavior of the device. The effects of these interactions needed to be understood in order for this task to be successful. Tasks 4 and 5 were devoted to production of devices using the novel technology developed in the previous tasks. In addition, characterization tests were required to ensure the devices would perform adequately as replacements for the existing technology.

Each of these tasks has been achieved successfully. In task 2, a series of potential materials were surveyed, and ranked in order of desirability. Prototype device structures were produced and characterized in order to do this. This satisfied the requirements for Task 2.

From the results of this relatively extensive survey, the number of candidate materials was reduced to one or two. Small devices were made in order to test the functionality of such samples, and a series of optimization experiments were carried out with encouraging results. Devices were fabricated, and some room temperature cycling carried out showing that there are no fundamental problems with this technology. This series of achievements satisfied the requirements for Tasks 3 and 4.

The results obtained from Task 3 naturally led to scale-up of the process, so a large cathode was obtained and installed in a spare slot in the production coater, and a series of large devices fabricated. In particular, devices with dimensions of 60” x 34” were produced, using processes which are fully compatible with mass production. Testing followed, satisfying the requirements for Task 5.

As can be seen from this discussion, all the requirements of the project have therefore been successfully achieved. The devices produced using the newly developed technology showed excellent optical properties, often exceeding the performance of the existing technology, equivalent durability results, and promise a significantly simplified manufacturing approach, thereby suggesting higher yields as a result of less handling, and therefore lower costs.

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Introduction

This report is divided into four main sections. The first introductory section deals briefly with background information intended to place the rest of the report in context. It includes a general introduction to electrochromic technology, followed by information on the tasks included in this project. There then follows a detailed description of work carried out on each of the tasks.

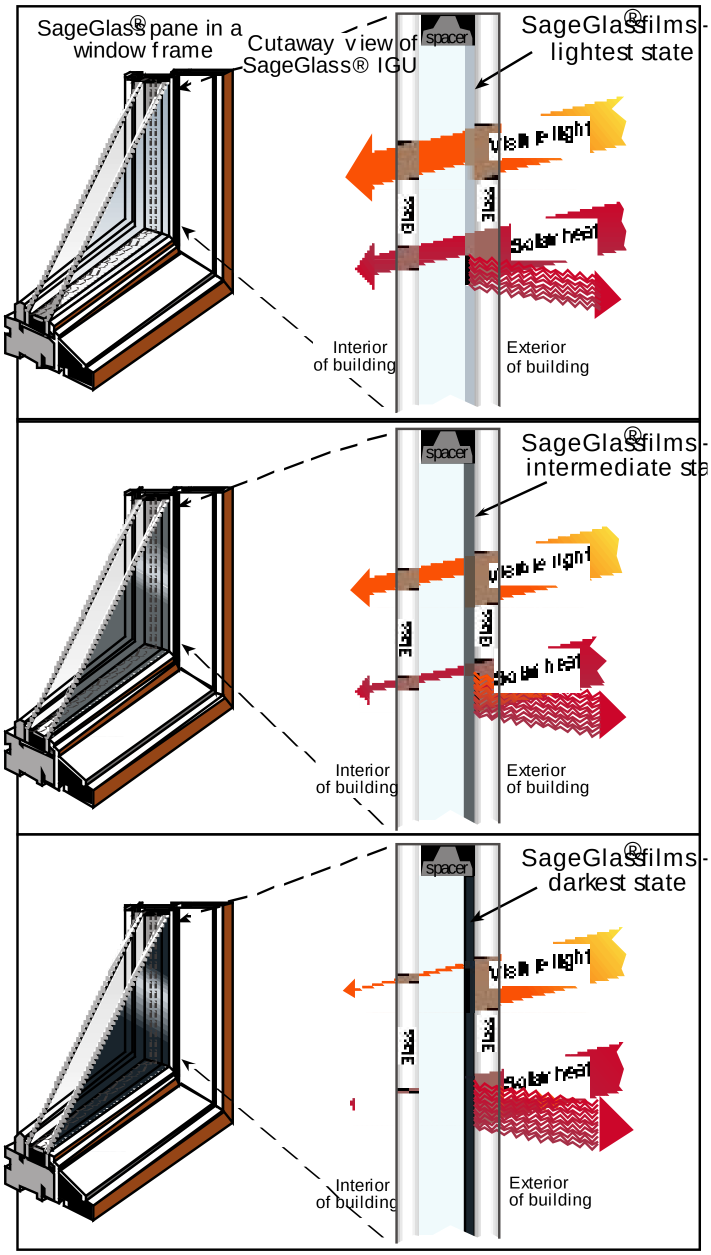

Figure 1 – Schematic of the principle of operation of a SageGlass EC window.

Description and Benefits of Electrochromic Windows

There is a need to improve the energy efficiency of building envelopes as they are the primary factor governing the heating, cooling, lighting and ventilation requirements of buildings – influencing 53% of building energy usage. In particular, windows contribute significantly to the overall energy performance of building envelopes, thus there is a need to develop advanced energy efficient window and glazing systems.

The electrochromic (EC) window represents the next generation of advanced glazing technology that will both reduce the energy consumed in buildings, and improve the overall comfort of the occupants. “Switchable” EC windows provide, on demand, dynamic control of visible light, solar heat gain, and glare without blocking the view. As exterior light levels change, the performance of the window can be electronically adjusted to suit conditions. A schematic illustrating how SageGlass® EC windows work is shown in Figure 1. In the clear, or untinted, state, the window is transparent to both the visible and the near infra-red radiation impinging on the window. This radiation is transmitted through the window into the building interior. The SageGlass® films can be tinted over the range of fully clear to fully colored, and held anywhere between these states. As the films are tinted, they become more absorbing, and depending on the level of tint, a fraction of the incident solar radiation is absorbed by the films. This reduces the amount of both visible and near-infra-red radiation transmitted through the window. The absorption of this energy causes the EC pane to heat up. As a result of the low-emissivity coatings, which are an intrinsic feature of the SageGlass® films, the heat built up in the EC pane is preferentially radiated back out of the building. This is also shown schematically in Figure 1. Notice that the visible transmission never drops to zero, so a view to the outside is always maintained, and after all, this is the reason for putting in a window in the first place.

SageGlass® EC glazings offer the potential to save cooling and lighting costs, with the added benefit of improving thermal and visual comfort. Control over solar heat gain will also result in the use of reduced capacity HVAC equipment.

If a step change in the energy efficiency and performance of buildings is to be achieved, there is a clear need to bring electrochromic technology to the marketplace. This project addressed the need to improve the manufacturing process by streamlining the process, thereby reducing the chances of introducing defects because of reduced handling, and thus increasing the yield. This will help to reduce the cost EC windows and help speed the introduction of EC windows into buildings and thus maximize the total energy savings in the US and worldwide.

The R&D activities in this project have resulted in a single in-line vacuum process for the production of EC window also resulting in better device performance and higher yields. The goal is to reduce the product costs to the point necessary for broad penetration of architectural markets.

EC Window Technology

The SAGE device is a series of thin films deposited onto a glass substrate one on top of the other to form a functional EC stack. The outermost layers are transparent conductors, which are used to apply a voltage to the active layers that are sandwiched between them. The active layers consist of an electrochromic (EC) layer, an ion conductor (IC) layer, and a counter electrode layer (CE). Electrical charge, in the form of electrons and ions, is shuttled between the CE and the EC layers, producing the bleached and colored states respectively: the electrons are passed around the outer circuit, while the ions are transported through the IC. Insertion of charge into the EC layer will cause that layer to color to a depth that depends upon the amount of charge transferred. The effect is completely reversible, and is accomplished simply by reversing the polarity of the voltage. This is shown schematically in Figure 2.

Significance of Problem

The U.S. Department of Energy has estimated that the use of EC windows could reduce peak electric loads in buildings by 20-30%.[1] Similarly, energy calculations comparing SageGlass® EC windows with commercially available static glazings in the Southwestern U.S. result in average energy savings of 28% for a cooling dominated environment. These and other simulations indicate that significant cumulative energy savings in the U.S. would be achieved if current static

[1] E.S. Lee *et al*. Energy Performance Analysis of Electrochromic Windows in New York Commercial Office Buildings, LBNL-50096, April 2002, Lawrence Berkeley National Laboratory, Berkeley, CA.

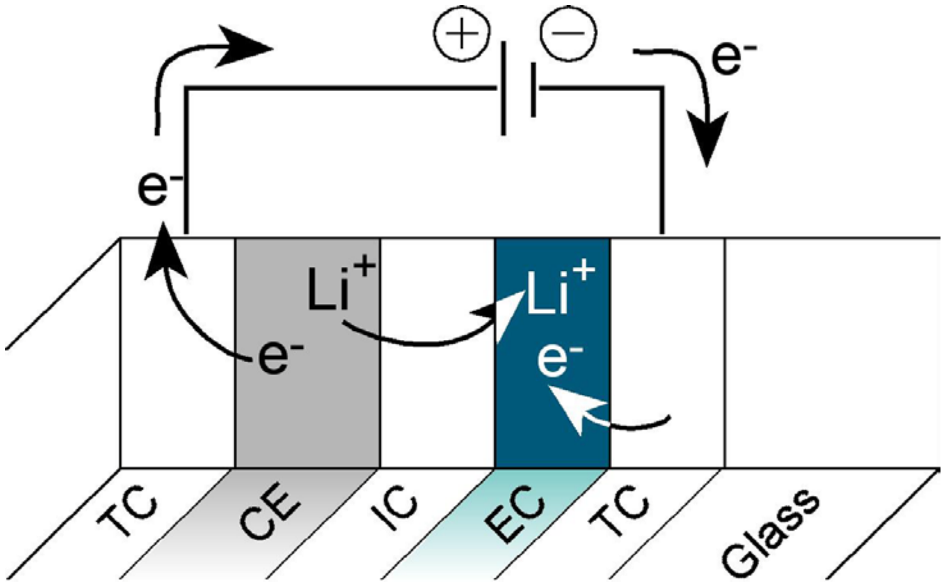

Figure 2 – The SageGlass® EC device showing the motion of the charged species around the 'circuit'.

glazings were replaced by dynamic EC windows in a significant portion of commercial and residential buildings.

EC windows can be lightened or darkened electrically to minimize solar heat gain and block glare while maintaining daylighting and a view through the window. They also significantly attenuate that part of the solar spectrum that causes fading of furniture and fabrics. This solid-state electrochromic window functions with no moving parts and can also provide security benefits since any break in the window can interrupt an electric circuit triggering an alarm.

To achieve significant national energy savings, the penetration of EC windows into existing markets must be maximized so that the largest cumulative energy reduction can be realized. The rapidity with which EC windows can be introduced and replace a current static glazing is a strong function of the cost. While the fundamental window performance requirements (e.g. optical quality, dynamic range, uniformity) could already be achieved, this proposal addressed the need to continually improve production efficiencies and throughput to achieve cost effective manufacturing.

High-performance low-e[2] is the standard for solar control windows today. This product is fabricated by continuous in-line sputter deposition of metal oxide materials on glass. The resulting commercial IGU price at production volumes of 50 million sq. ft. per year is $6 to $20 per sq.ft.[2] The SAGE coating process is essentially the same with the notable exception that the ion conductor (IC) layer is deposited by solution coating. This necessitates interrupting the vacuum coating process to solution-coat and heat the IC layer. Removing this costly and time-consuming step would significantly reduce the cost of the EC IGU. Figure 3 shows the current process compared with the desired process.

The problem is to replace the two-coater batch process shown in the upper portion of Figure 3 with a continuous in-line vacuum process. This requires the development of a robust, fully functional sputtered ion conductor to replace the current IC thin-film. The goal is to bring the volume price of an EC IGU to within a factor of two of the current low-e product price. Today, the SAGE process involves vacuum deposition of the first coatings, followed by a solution

[2] Major commercial glass fabricator

coating step, and then a final vacuum deposition. This has a significant impact on operations and yields. The following factors contribute to the increased costs of solution coating over a continuous in-line process:

- Increased cost of two coaters (versus one) and, equally important, the additional time required to transport partially completed EC devices from one coater to another;
- Capital costs for precision firing furnace(s) and expensive carrier fixtures. These costs have significant impact on product price throughout the production ramp-up;
- Costs of solutions, labor, and facilities operations – operators are required to move glass through dipping and firing steps, which must be carried out in a high-overhead class 1000 clean room;
- Hazardous waste (from dipping solution) that must be monitored and disposed of properly. There are no other volatile organic compounds (VOCs) in our process.
- Reduced yields due to handling and particulates that can accumulate on surfaces from successive vacuum bleed-up, dipping, and pump down.

There is a lost opportunity cost. As the glass volume increases the number of ovens, dippers, and the clean room area also increase. This will force us to incur facility expansion and additional overhead costs much earlier than would be required for an all-vacuum process.

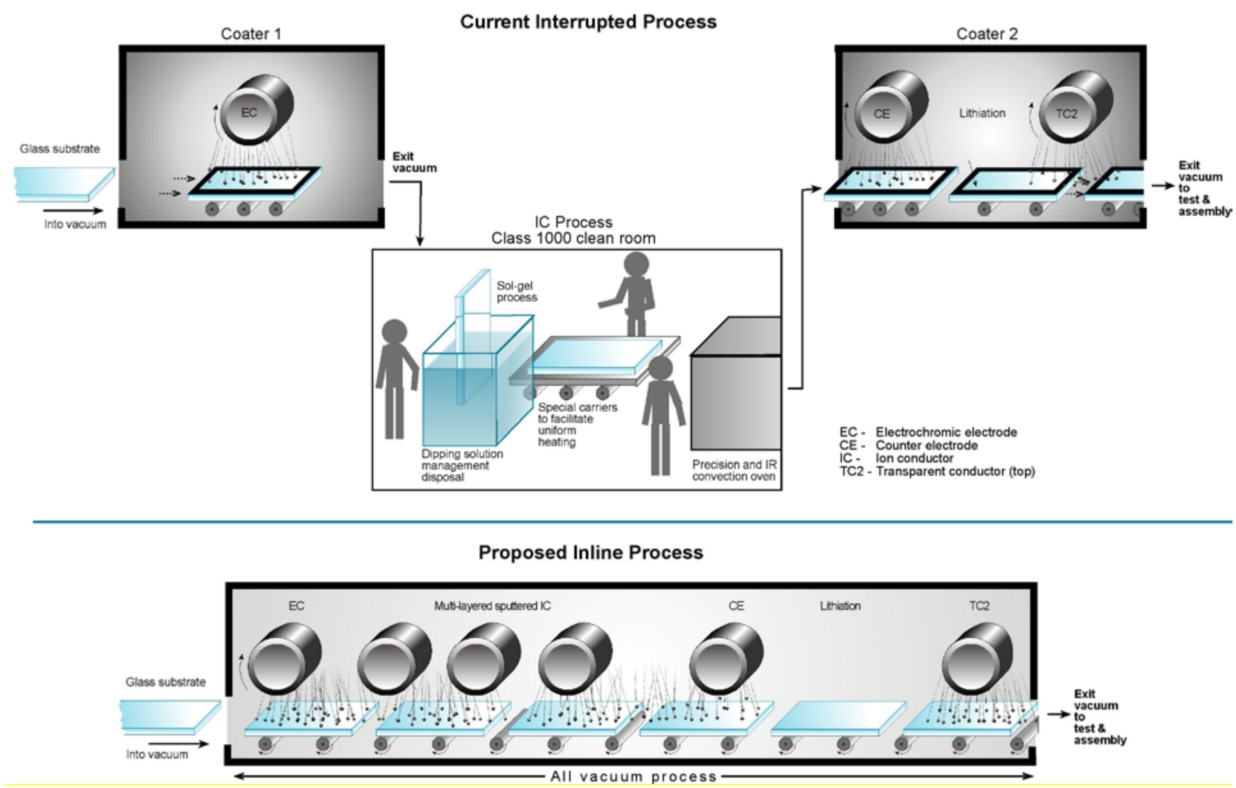

Figure 3 – The current process for depositing the EC coatings is interrupted by the IC dipping step, which is time-consuming, inefficient, costly, and invites opportunity for yield loss through handling and exposure to air, is compared to the envisioned inline process, all of which takes place inside vacuum chambers.

Developing an all sputtered thin-film ion conductor is a challenging technology project and has a number of associated technological risks:

(1) The IC must conduct ions, serve as a barrier to electrons, and yield uniform device coloration. The challenge is to develop sputtered IC technology that allows good ionic transport for rapid switching with very low electronic leakage.

(2) The IC must be stable to long term solar exposure, large changes in temperature, repetitive thermal and potential cycling, and over-voltages.[3]

(3) Optical indices of the materials need to be matched to the surrounding layers to minimize reflections and the resulting attenuation of device dynamic range.

(4) Deposition rates need to be high enough for economical processing.

Summary

This project was focused on replacing one particular aspect of production currently carried out using a non-vacuum process, to result in a single in-line continuous vacuum process where raw glass is fed into the system at one end, and completed EC windows exit at the other.

Previous attempts to do this have resulted in either weakly-coloring, electronically-leaky devices, or devices which colored extremely slowly or not at all. The causes for this were not completely clear at the outset of the project, and so the work program was designed to allow investigation of multiple technology aspects while driving towards a manufacturing solution.

This report discusses the activities carried out to achieve this goal.

[3] R.B. Goldner et al. "Electrochromic window with high reflectivity modulation", US. Patent 6,094,292 Jul 25, 2000.

Report on Work Plan Activities

The following is a detailed description of the technical progress achieved.

Task 1. Develop All Vacuum Deposited Electrochromic (EC) Device

Previous SageGlass® products have used a non-vacuum process for the IC layer. In order for the EC glass to be processed, the glass has to undergo two separate vacuum process steps, separated by the IC deposition process. Naturally, this entails cycling the substrates into and out of the vacuum systems through load locks, thereby increasing the potential contamination introduced onto the substrate. This is undesirable, and so this section outlines the work done during this project to modify the process to allow the IC layer to be deposited during a continuous in-line vacuum process thereby eliminate several potential contamination sources, as well as simplifying the process considerably.

Task 1.1 Carry out Baseline Experiments

Some initial experiments to prove the feasibility were carried out using a conveniently available material, which was not necessarily ideal for the final product, as it was known to have a lower than desired refractive index. A target of the material was, however, already present in the coater, and so many of the initial screening experiments could be carried out conveniently with it.

These experiments involved range-finding for the thickness, sputtering conditions and heating processes. The general idea was to find the trends associated with changes to the process, rather than to zero in on a particular device structure. Later experiments would be used to narrow down the process window, both in terms of materials and process parameters.

Initially, a single layer was used as a direct drop-in replacement of the proprietary non-sputtered IC. This was used to produce a number of devices, and some limited optimization experiments were carried out with a view to understanding the most significant experimental factors.

Several factorial experiments were carried out on relatively large area substrates. These revealed some dependence on the factors which are known to be important to 'normal' device behavior. Indeed, we were able to produce some 18"x42" samples that showed good coloration (down to around 5%, compared with the more normal 2%) but at slightly elevated leakage currents. Attempts to produce further samples at the same conditions resulted in somewhat mixed results, suggesting there was at least one other factor which was not being controlled closely enough for stable process yields. Further investigation of the process sensitivity to the most likely experimental factors was required and undertaken, and discussed later in this report.

An additional effort which has been carried out with respect to the measurement of these characteristics is to develop an instrument capable of measuring the electrical and optical characteristics of small elements of the device, which have previously been isolated physically by a proprietary laser scribing technique. By sampling small areas, it is possible to eliminate the effect of the sheet resistance of the transparent conductors, and thereby characterize the film stack at that particular location. This further allows any non-uniformities due to film processing to be investigated.

Task 1.2 Design Screening Experiment

DOE (Design of Experiments) techniques were used to achieve the maximum access to process parameter space in terms of IC materials combinations offering the potential for high performance. The multi-layer combinations were selected primarily to set up barriers to electronic current while retaining ionic conductivity.

In order to carry out these high throughput screening experiments, it was necessary to refurbish the Laboratory Coater in order to be able to utilize the Production Coater most efficiently. This is due to both the relatively high cost for large targets, and the loss of valuable production time while the experiments are being carried out.

This means that the Production Coater can be used to deposit 'standard' Coat1 films, i.e. the electrochromic (EC) layers. Then the Lab Coater is used to deposit a variety of sputtered ion conductor(IC) layers, before the Production Coater is used to finish off the devices. Because the lab coater can only coat a relatively small area, the Production Coater can be used to coat a large number of samples in a single carrier. The additional advantage is that the coatings from the production coater will be extremely uniform.

The benefit of using the laboratory coater for depositing the IC layers can be further leveraged when the target is modified by clamping or bonding different materials to it. This permits exploration of different compositions by variation of the relative amounts of the two (or more) materials, even within the same substrate. The photograph in Figure 4 shows an example of a cathode set-up to allow this. The diagonal strips are a different material to the base target, and are held on by a clamping arrangement. Significant work was necessary to design and fabricate the novel target arrangement, and this was carried out successfully in-house. One of the issues was ensuring that the metal strips remain in good thermal contact with the cathode to prevent warping, and subsequent variable deposition rates as a result of uneven heating.

Figure 4 – Photograph showing the method of producing a composite target by clamping strips of one material to a target of another.

The first series of experiments were designed to screen a variety of potential IC materials. It has been found that the best way to carry out these types of material evaluations is to fabricate complete devices, varying some relevant parameters in controlled ways as part of designed experiments. In this case, we varied the IC material, whilst keeping as many of the other parameters as possible constant. In this way, direct comparisons between the properties obtained using different materials are possible, and therefore a desirability ranking can be established.

Task 1.3 Evaluate Multilayer

Given the limited but encouraging results from the initial investigation of a single layer IC of Material A, it was prudent to suspect that the simple system may not have a wide enough process window to enable a viable process to operate for this particular IC material. To anticipate this, a series of experiments were begun to investigate a multilayer ion conductor. This system is intended to increase the electronic impedance of the overall IC layer, whilst maintaining high ionic conductivity.

Early proof-of-concept work for this system indicated that the electronic breakdown voltage[4] of the system is increased, without significantly impacting the ionic dynamics. This is important because it prevents additional electronic current from flowing through the transparent conductors, thereby dropping voltage across the device. (Remember, the coloration that a particular element of the device reaches depends on the voltage applied across the element – any voltage drop through the transparent conductors will change the voltage drops across the device, leading to non-uniform coloration.)

Experiments were carried out with a triple layer where Material A was deposited on either side of a layer of the EC material to form the multilayer IC. Several different arrangements of the multilayer IC were attempted, but with the existing materials, it was found that the increase in electronic breakdown voltage was not as much as had been hoped for. This approach was therefore placed on hold.

Task 2. Perform High Throughput Screening Experiments

This section discusses the high throughput screening experiments carried out as part of the project. The process has been described in the previous section. The objective is to identify key replacement materials, presumably metal oxides, which have the required electrical, ionic transport and optical characteristics to be capable of replacing the existing material. In addition, the material selected should be capable of being fabricated into a usable sputtering target, meaning it is not too brittle, moisture sensitive or hazardous, to be useful as a production material. Furthermore, the chosen material should be reasonably priced, as the overall aim of the project is to reduce the cost of the finished product, so choosing a material which is prohibitively expensive runs counter to this objective.

The IC material must be ionically conducting and electronically insulating, transparent, and preferably have a refractive index close to that of the surrounding layers to minimize reflections. Several different materials were investigated, and were chosen for one or more of a variety of reasons. First, from practical experimentation, we have found that there are materials, which although not considered 'classical' ionic conductors, can be used as the IC layer in an EC device by virtue of being thin enough to allow passage of ions while preventing the transmission of electrons. Next, materials were considered which were well-known as ion conductors. In addition, we considered forming composite materials of higher index materials with materials which either had good electronic characteristics, or good ion transport properties. Obviously, using these criteria will result in a huge number of potential material combinations, so it was necessary to limit the numbers, which we did by considering the ease of availability and the cost

[4] Previous work has shown that the electronic behavior of an element of an all-solid state EC device shows distinctive non-linear behavior. In the idealized form this behavior takes the form of a very high resistance region with a low slope, up to a certain voltage (referred to as the threshold voltage), whereupon it begins to pass significant amounts of current, as if the device were a forward biased diode. The threshold voltage is typically around 2V in 'standard' devices. Any increase in the electronic breakdown will have a beneficial effect, from the perspective of yielding more uniform coloration, and also consuming less power as a result of leakage.

amongst other parameters. We eventually arrived at a short list of around eight materials which were subjected to the initial screening.

Material A was one which was already the basis of the current standard IC layer. Material B was chosen as it is a similar material to Material A, providing a good electrical barrier, but when deposited thin enough can allow diffuse of the ions through it. Material C was chosen as it has a higher refractive index, comparable to those expected from surrounding layers. Material D is a mixture of materials A and B. The remaining material combinations – E to H – are chosen because of refractive index considerations, and are all mixtures.

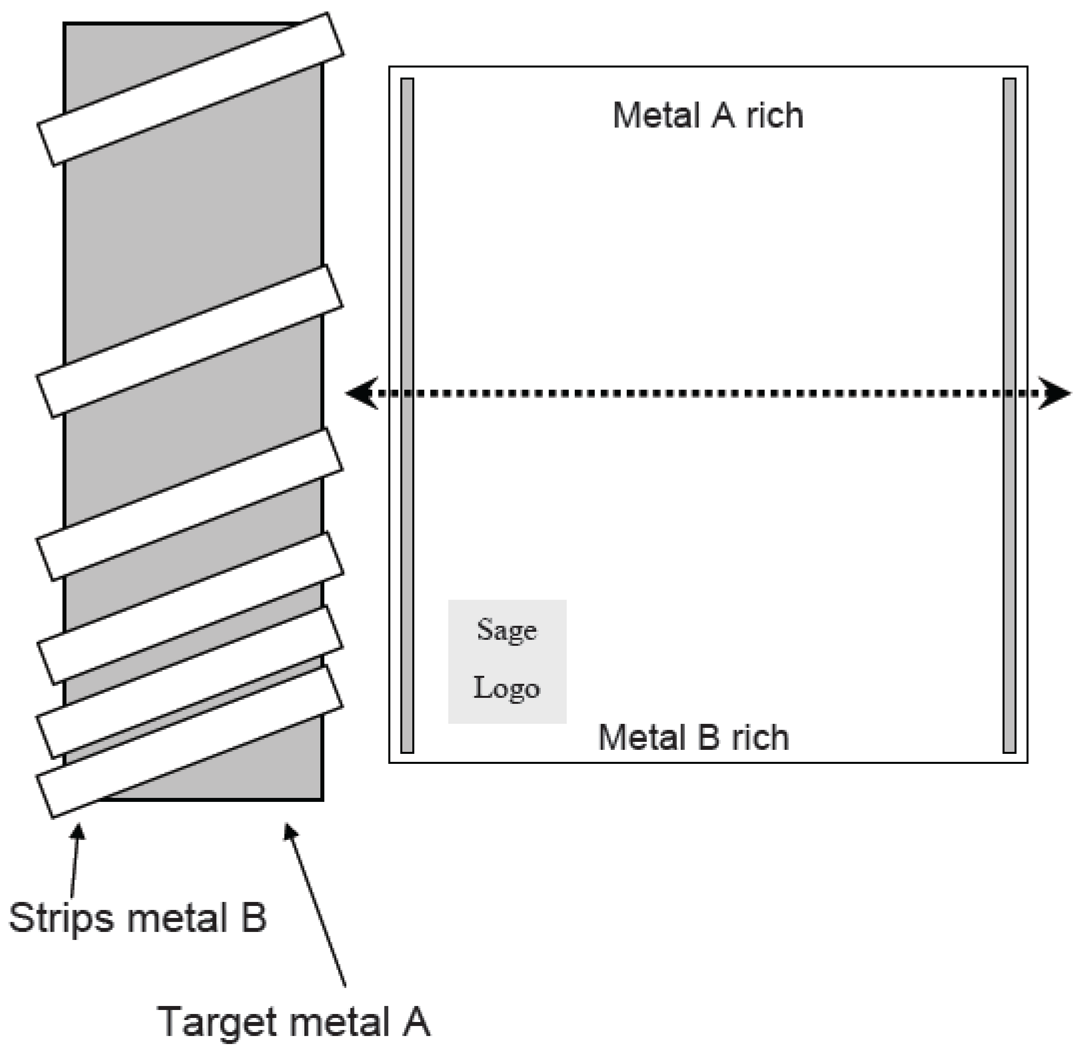

Figure 5 – Layout of the metal strips of material A (for example) upon the target of metal B. The non-uniform placing results in a composition gradient on the substrate. The arrow indicates the direction of travel of the substrate across the target.

In this series of experiments, the EC layer was deposited onto a 'standard' substrate using the standard production process on the production coater. The experimental IC was then deposited onto this layer using the lab coater, before the remaining layers were deposited on the production coater.

During the initial screening work we estimated the two major parameters most likely to strongly influence the EC device behavior, and then conducted a limited set of experiments to identify the most suitable materials for further study. Each of the materials was deposited with three different thicknesses at two different sputter pressures. In addition, an attempt to investigate the composition of the material system was made by depositing the mixed metal from a target with a non-uniform distribution of metal strips as shown in Figure 5.

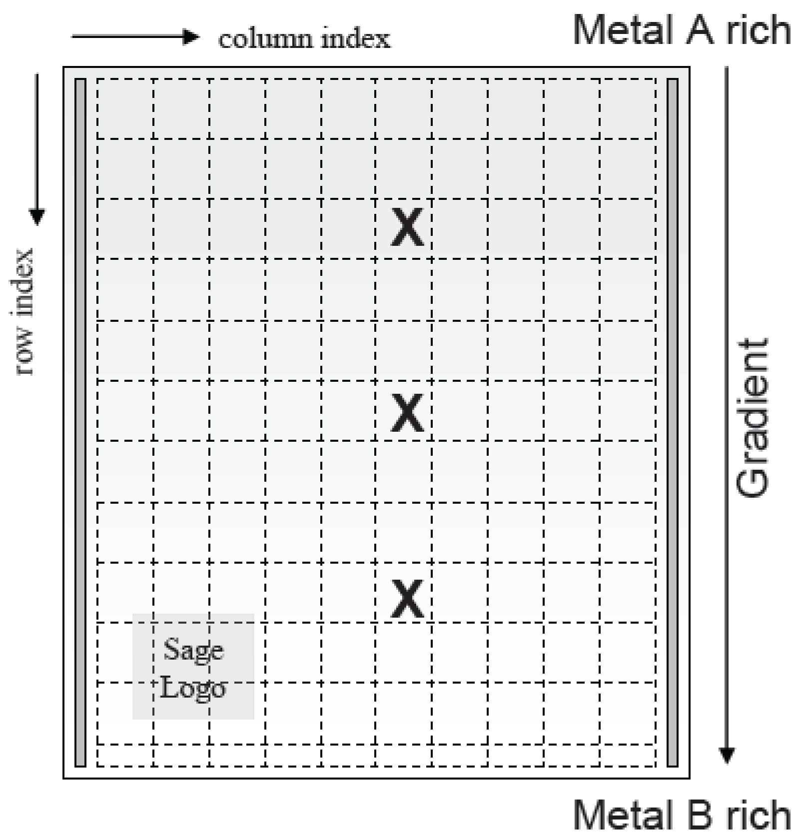

Figure 6 – Layout of the selective laser scribing showing the bus bars and laser scribes.

Thick samples were deposited onto silicon wafers in order to determine the thickness and hence the deposition rate by depositing under the relevant conditions for an appropriate length of time. The samples were sputtered in a reactive environment using argon and oxygen with sufficient oxygen to yield fully oxidized films.

Several other parameters were ignored during this initial study, such as oxygen content, substrate temperature, deposition power, and any other modification to the other layers in the stack, as this would have increased the number of samples dramatically.

Following deposition, selective laser scribing was carried out to divide the samples into pixels which could be individually switched and characterized. Electrical contact was made to the individual pixels using conductive copper tape. This is shown schematically in Figure 6. For a mixed metal sample, three different compositions were measured to obtain information characteristic of three different compositions. For single oxides three pixels were also measured to give a better understanding of the variation in the measurement.

A standard test is carried out on each pixel. This consists of applying a constant (positive) coloring voltage to the pixel to color it, followed by a constant (negative) voltage to clear it. The current is measured using an in-line ammeter, and the optical properties are measured using a fiber-optic based spectrometer system, which allows the measurement to be localized to the pixel of interest.

The data used to characterize the pixel are derived from measurement of the optical properties, and the electrical current flowing through the pixel as a function of time. The two optical characteristics were (1) the coloration rate, or switching speed, derived from the contrast ratio (i.e. the initial transmission/instantaneous transmission) and (2) the transmission in the colored and cleared states. The validity of using the contrast ratio to measure coloration rate derives from the Beer-Lambert law, as the gradient of the contrast ratio is constant for the duration of the coloration. In this case, we analyzed all the optical data at a characteristic wavelength of 550nm.

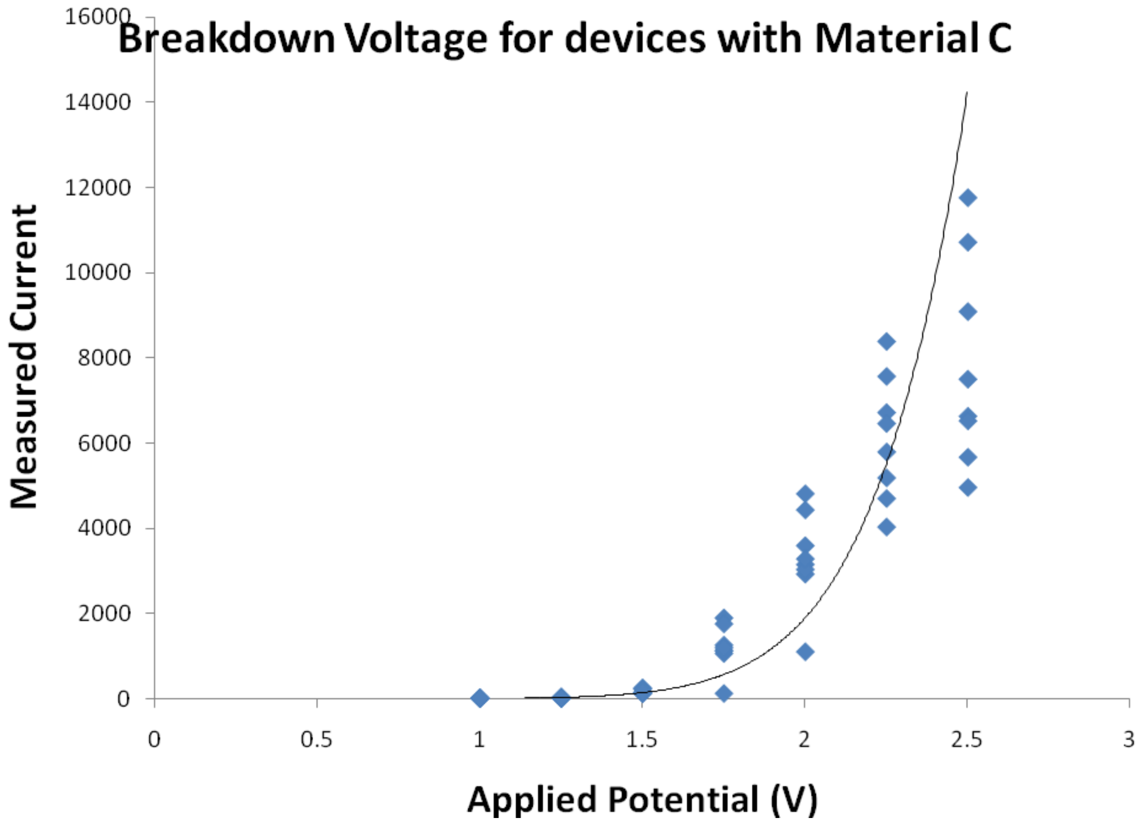

Figure 7 – A plot of the saturation current as a function of the applied voltage for several devices with Material C as the IC material.

The electrical measurements were taken by stepping the voltage from zero volts to 4V in steps of 0.2V, allowing the current to stabilize at each stepped voltage. The saturation current at each voltage is then plotted to allow determination of the coloration voltage. We have found in previous work that the electrical characteristic of an EC device resembles a diode with a turn-on voltage of a few volts. Generally speaking, a 'good' EC device has a very low intrinsic – i.e. not associated with extrinsic defects – leakage current up to the point at which the 'diode' breaks down. The current through the device above the breakdown voltage is then determined by the electrical resistance of the Ohmic portions of the device, i.e. the sheet resistance of the transparent electronic conductors. Obviously higher breakdown voltage is better.

A typical result is shown in Figure 7 for Material C of a certain composition. The breakdown voltage is determined by fitting a line to the plot at higher voltages, and projecting back to the voltage axis.

To summarize, a significant number of samples were prepared and their performance characteristics measured. Each of the eight materials were prepared at 3 different thicknesses, at two different sputter powers (and hence thickness). Five of the composite samples were then measured at three different compositions.

Results

Material A showed a low contrast ratio, and slow coloration rate, with the coloration rate increasing with decreasing sputter power. This is somewhat significant as the present material shows much better performance, and is nominally the same composition.

Material B shows a relatively high coloration rate, with no apparent variation with layer thickness. The clear state transmission decreased as a function of layer thickness, due to optical interference between the relatively low refractive index of the IC layer and the surrounding layers.

Material C showed almost no fringes in the clear state, showing a good optical match between the IC and EC layers as required, and this leads to a relatively high value of the clear state transmission of around 75%, independent of the IC thickness. The coloration rate is relatively high, but a little lower than that seen for Material B. The coloration rate was seen to increase with increasing thickness at the lower sputter pressure, but decrease with thickness at the higher sputter pressure. The coloration rate was seen to increase as the sputter pressure increased.

Material D showed a significant interference effect between the IC and the surrounding layers, which increased with increasing IC layer thickness, and this in turn caused a decrease in the clear state transmission, from 72% to around 65% for the thickest layer. Material D showed a moderate coloration rate, lower than both Material C and Material D.

Material E showed interference fringing intermediate between that of Material B and C. The visible light transmission remained fairly high – between 72 and 78% - and the coloration rate was relatively high. The coloration rate was found to decrease with increasing layer thickness, while increasing with increasing sputter pressure. In addition, a dependence of coloration rate was found on the composition, but this was significantly lower than the effect of sputter pressure and thickness.

Material F showed minor fringing in the clear state reflecting a good optical match yielding a high clear state transmission. The coloration rate was relatively slow, approximately a factor of two slower than Materials B and C. The coloration rate was seen to decrease with increasing layer thickness, but increase with increasing sputter pressure. Once again, a variation of coloration rate was seen with composition.

Material G showed significant fringing in the clear state when compared to the other mixed metal oxide systems. Therefore, the clear state transmission was moderate, being somewhere between 65 and 70%. The coloration rate was relatively slow, and decreased with increasing thickness, but increased with increasing sputter pressure. There was also a dependence of the coloration rate on composition, as seen with some of the other mixed systems.

Material H showed minor fringing in the clear state, a moderate clear state and a slow coloration rate. There was a large degree of scatter in the results, throwing the conclusions into question, however, there was little impact of the thickness on coloration rate, but the rate appeared to increase with increasing thickness.

Figure 8 shows representative measurements of the transmission change with time for each of the materials, and a characteristic of a standard device for comparison. It can be seen that some of the materials (B, C and E) appear to be relatively promising, showing good switching speed and contrast ratio, although they are not quite yet as good as the standard device.

Electrical data such as shown in Figure 7 was obtained for all the materials except Materials G and H and the breakdown voltage extracted. .These results are shown in Figure 9. It is clear that the breakdown voltages for Materials E and F appear to be slightly better than the others.

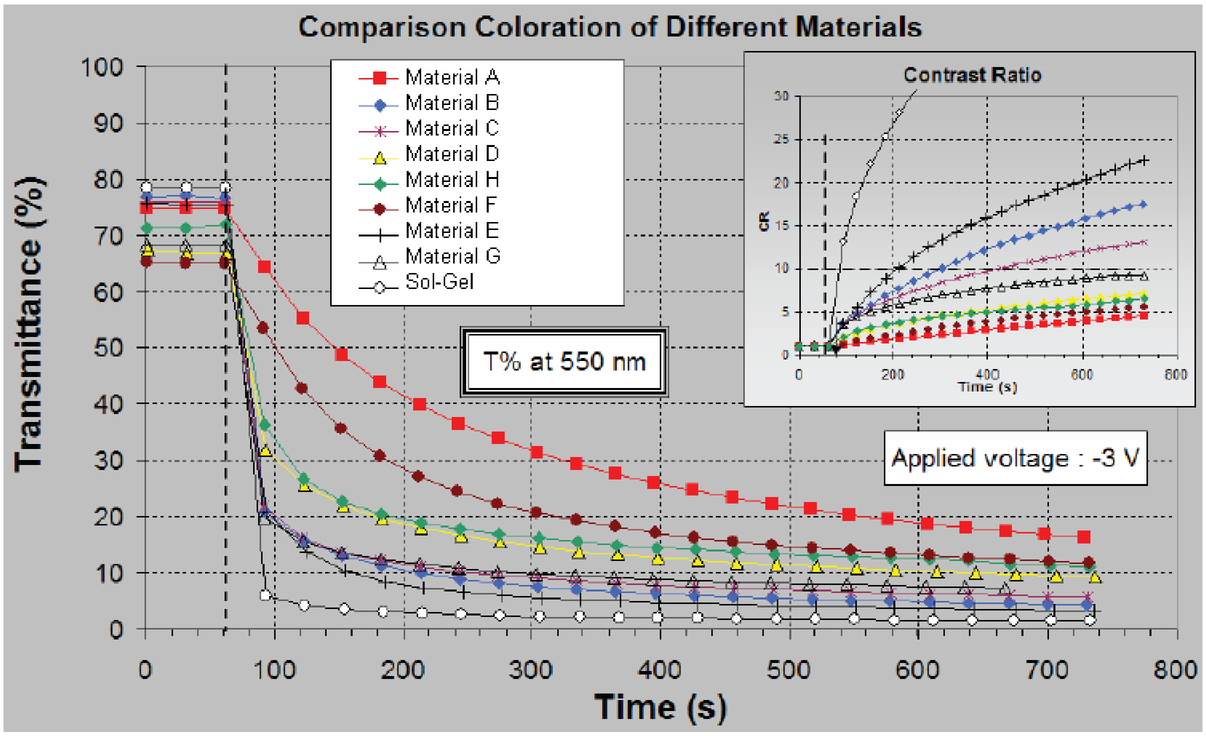

Figure 8 – Representative transmission versus time curves from each material as well as a comparison curve from a standard device. There is a big difference between the materials, and Materials E, B and C appear to be the most promising.

Summary

It can be seen that for each of the measurements – optical matching, electrical, switching speed – no one material is best. The choice must therefore be a compromise based on the results of each of the experiments. For example, Material B shows good kinetics, but has a relatively low refractive index. Material C has excellent optical indices, but is slower to color that Material B. The mixture of B and C (Material H) does not seem to offer an advantage over either. Material E seems to be the most promising, with the highest breakdown voltage, fastest coloration rate, but it was found that the optimum thickness was very thin and could be problematical for process control, as it may be difficult to measure in-situ.

Although Material E seemed to be a slightly better candidate, the initial enquiries into target manufacture suggested it may be difficult to make into a target. In order to proceed with more detailed optimization experiments in Task 2, Material C was selected as the initial candidate, as it was readily available, relatively inexpensive, easy to sputter and has reasonable properties as determined by the experiments described above.

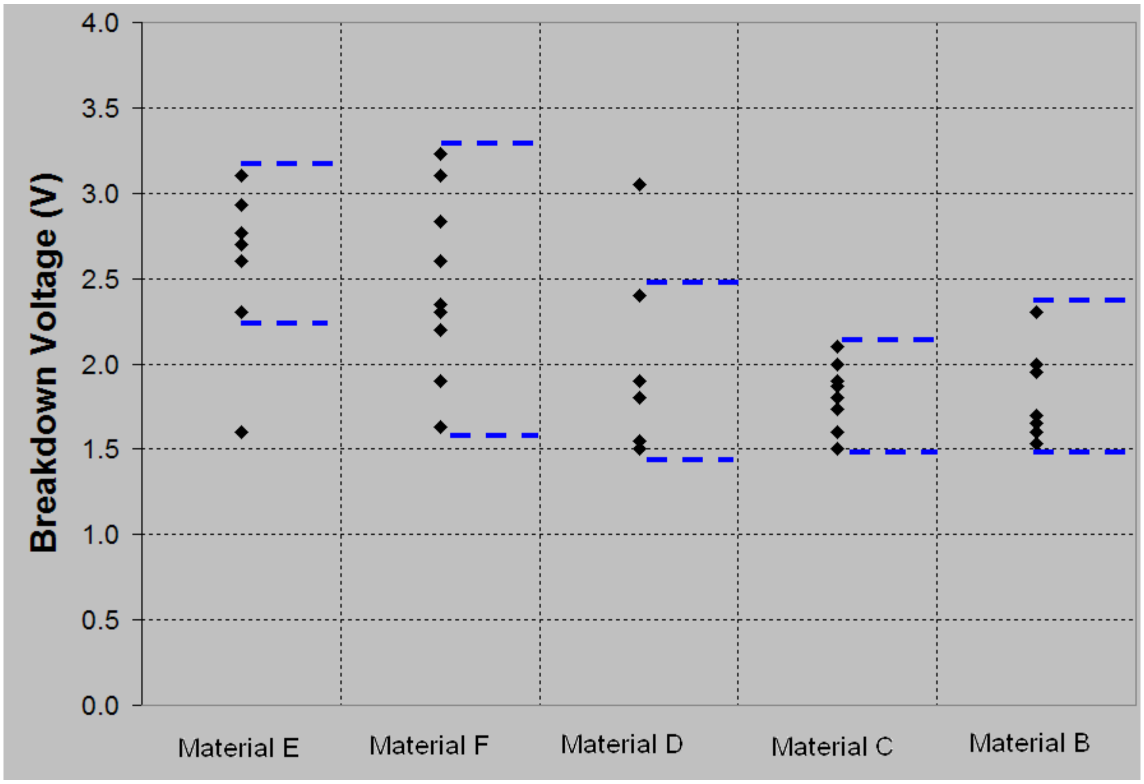

Figure 9 – Measured breakdown voltages from the experiments discussed in the text.

Task 3. Carry Out Optimization Experiments

The aim of this task was to select no more than two likely materials combinations and carry out optimization experiments. Material C was selected as it was readily available and offered promising performance. A large rotary target was obtained and loaded into the Production Coating equipment.

The optimization experiments needed to encompass several aspects of device development: including the substrate, and also the layer thicknesses, heating requirements and sputtering process parameters. Some of the experiments carried out along these lines are described in the following section.

Task 3.1 DOE Process Optimization

In this section, specific experiments will be discussed which utilized Design of Experiments to optimize process parameters (thickness, heat treatment, number of layers, etc.). The response variables which are used to judge the success of the optimization are the usual electrochromic parameters including leakage current, coloration time, dark state transmission, etc.

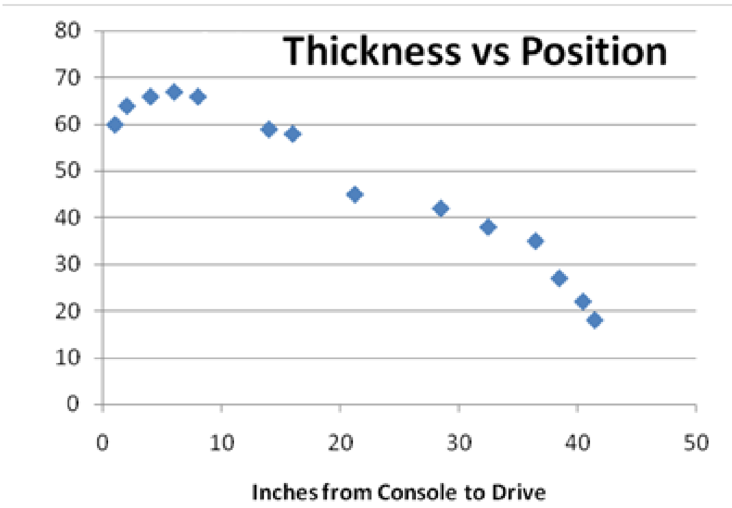

Figure 10 – Thickness of IC layer as a function of position across the deposition zone for a test deposition.

One of the first experiments was designed to look at the dependence of the device behavior on the IC thickness. A physical mask was installed along with the sputter target. Witness strips were then run through to give an indication of the thickness. Work was also needed to establish conditions for the IC deposition. Baseline deposition conditions were established with a relatively high percentage of oxygen in the sputter gas, moderately high pressure and moderate power.

Figure 10 shows a plot of the relative thickness of the IC as a function of the position across the coating zone. This film was deposited in a regime where the deposition rate was approximately linear with deposition power, and so allowed the calculation of the required thickness for subsequent experiments.

Using the physical mask, we were able to produce samples with a variation in the IC thickness across the width of the devices. In addition a novel treatment was also varied across a number of devices, which are shown in Figure 11. They were isolated into strips perpendicular to the IC thickness variation, using a laser, and then tested. The thin IC is towards the top of the picture, and as can be seen the strips switch to various depths of coloration. Certain strips where there is an electrical shorting defect somewhere along its length are held almost entirely transparent, and these can be discounted as far as evaluating the intrinsic behavior of the film stack.

Reasonable behavior is seen, particularly in the device on the extreme right in Figure 11, although the overall currents for these devices were high, but this is not surprising, as there are a number of thin strips which are likely to contribute greatly to the leakage current.

We were able to use the results gleaned from these devices to generate a further DOE based on two IC thickness levels and several other experimentally variable factors. This was a five factor two-level experiment designed and carried out on a series of 10”x12” sized substrates. The variable factors were (a) IC deposition power, (b) IC deposition pressure, (c) novel treatment, (d) level of dopant and (e) IC thermal processing. In addition, samples were deposited on two

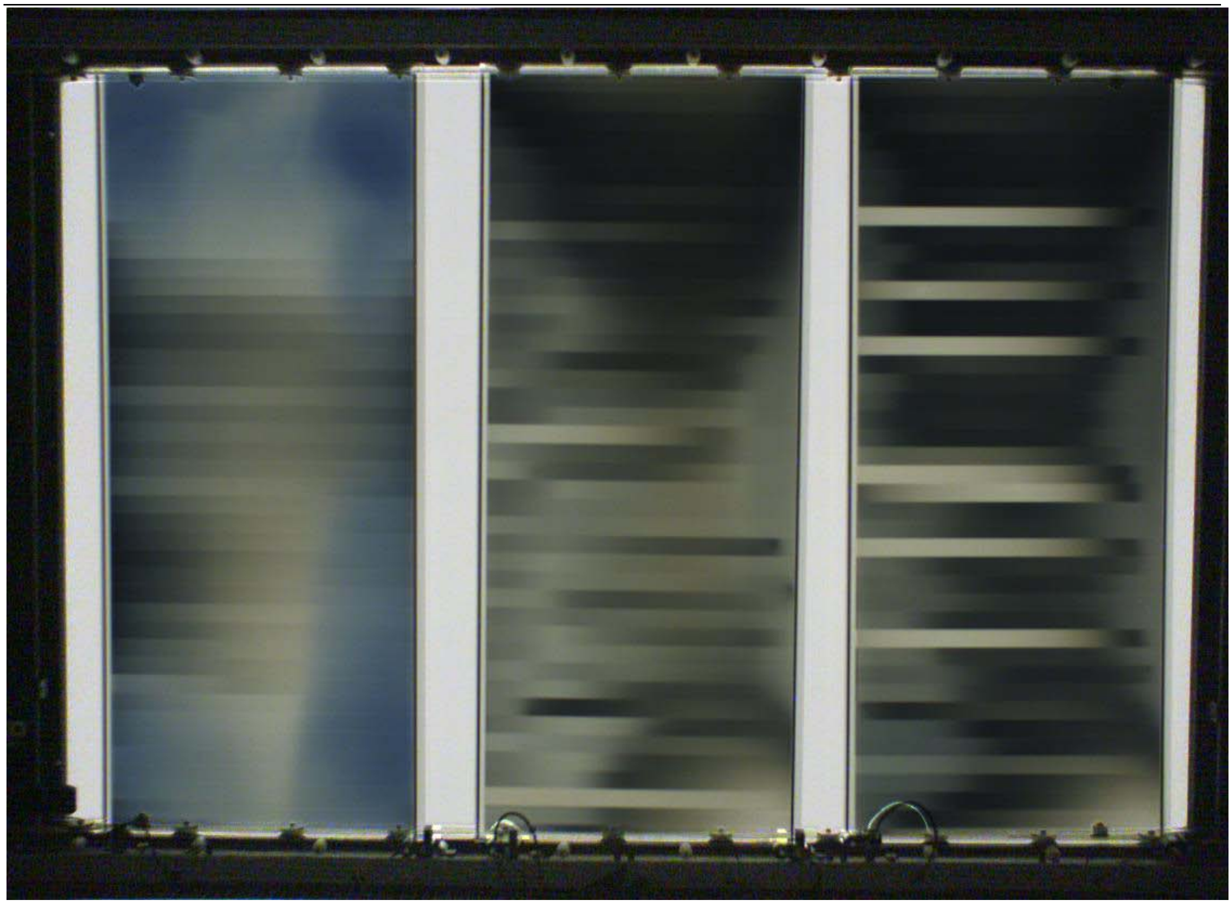

Figure 11 – Devices with a variation in the IC thickness (thicker at the bottom of the picture) in the colored state. These devices are laser isolated into strips perpendicular to the thickness variation.

different transparent conductive materials as part of the same experiment.

Several of the devices produced switched very well – over a wide dynamic range (in some cases with a ratio of T(bleach):T(color) of over 20) and at a rate which is similar to current technology. The ‘best’ device was measured to have a range of 73.9% to 3.6% with a low leakage current and colored to OD=1 (equivalent to 7.4%) in 58s.

Significant dependences of key parameters were identified. These were very nicely delineated, with the data showing that the differences between replicates (i.e. pure error) were very small compared with the significant effects identified. Most of the dependencies identified correlate well with our present understanding, for example, the dopant levels were responsible for the coloration depth, leakage currents, switching speed, etc. as we have seen before. Overall, this experiment provided an excellent base for performing further experiments to improve our understanding, and map out a possible production process.

Interestingly, in this experiment, the IC thickness (power) did not appear to impact the switching speed, but it appeared to reduce the leakage current as the power was increased. The IC deposition pressure did not appear to have any impact on any of the factors measured here, within the range we investigated, where both levels could probably be considered to be ‘high’.

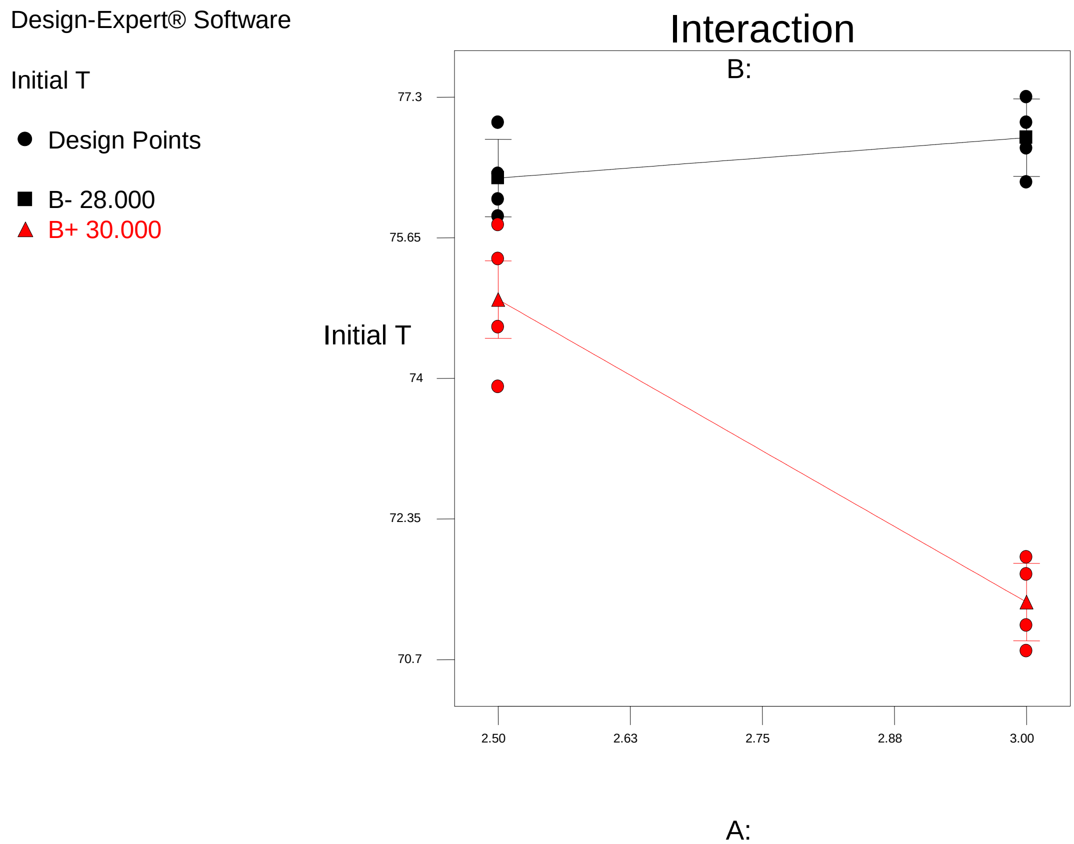

Figure 12 – Results of the measurement of the Initial transmission as a function of two of the experimental factors showing the excellent discrimination between the signal and the noise.

One welcome result was that devices deposited on the alternative transparent conductor showed good switching ranges. Where data were obtained, few differences between this and standard TCO samples were detected. Further investigation is warranted, especially in attempting to understand the requirements for any barrier layers or anti-reflection layers in terms of optical performance and film adhesion.

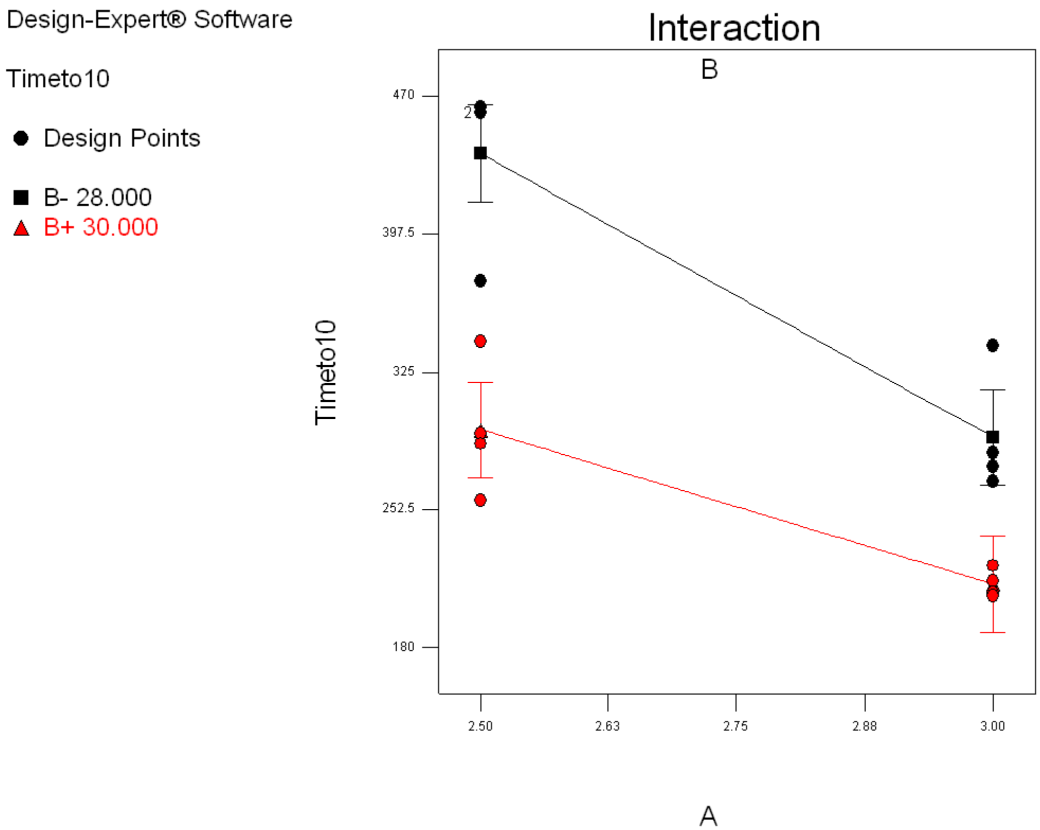

Figure 13 – Results of the measurements of the time taken to color to a transmission of 10%for the experiment discussed in the text.

Although there were several additional DOE's which have contributed further understanding to some of the details of the processing and layer structure, one additional experiment yielded devices showing properties very close to those required, and that included similar experimental factors to the previous experiment, with the addition of the thickness of certain critical layers. Once again, the results of the characterization measurements showed extremely good resolution between the effects and the pure noise. As an example, Figure 12 shows the results of the measurement of the initial transmission as a function of two of the variable factors. The grouping of the measurements in Figure 12 shows that the experiment was very well carried out and the results likely to be highly valid.

In fact, even a measurement of the coloring time showed good discrimination, as shown in Figure 13, and this is a measurement which is subject to several extrinsic influences, and so is usually significantly more noisy than the data seen here.

Table 1 summarizes the average performance of the sputtered IC devices and an average of a significant number (around 30) of recent production devices, by directly comparing critical measured parameters between the two sets of devices. The number of sputtered IC devices used to generate these data is approximately 15-20, but it is to be understood that these devices are generated as a result of process optimization experiments, so all are intentionally slightly different from each other. This comparison is simply intended to show that the new process is quite close to the existing production process, and it can easily be seen that further optimization experiments should bring all the measured parameters directly into line with, if not better than, the existing production.

These devices are all 18” x 18” (45.7cm x 45.7cm) in size. The comparison is very interesting, in that the majority of the parameters for the sputtered IC are very close to the existing production measurements. It should be pointed out that the measurements for the new sputtered IC devices are averaged over many samples, and that the ‘best’ devices exceed this average performance, coming very close to the behavior of the production samples. Note that in this experiment, the AR capping layer was omitted for the sputtered IC devices – this enables some diagnostic measurement to be taken on the device which would not be possible with the insulating capping layer present – and this would lead to an increase of a few percent in the initial transmission, bringing it directly in line with the production devices. Further subtle optimization of layer thicknesses should bring the leakage currents and switching speeds fully into line too.

Property	Average of recent sol-gel devices	New Sputtered IC
Initial T (%)	76.7	74.9
Dark T (%)	1.8	2.3
Bleach T (%)	73.9	71.9
Time to 10% (s)	360	429

Table 1 – Comparison between measured properties of sol-gel production devices with devices deposited with sputtered IC comprising Material C for devices of the same size.

To summarize, the goal of Task 2.1 has been achieved, as a process has been identified and several important key parameters optimized to produce devices with performance characteristics close to the existing production process. Critical process parameters have also been identified which will enable further improvements in the device behavior.

Task 3.2 Maximize Optical Transmission

The objective of this task was to ensure that the optical behavior of the sputtered IC was optimized to ensure maximum optical transparency. This would have been particularly important in the case of the multilayer IC which was proposed and investigated in the previous task. So far, that option has not been pursued thoroughly, but the modeling done to date indicates that by simply increasing the refractive index of the IC layer, there is a significant improvement in the clear state transmission and a reduction in the intensity of the reflected color.

In order to model the optical performance of the device it was necessary to measure the transmission and reflection of representative single films. There was an opportunity to construct a spectrophotometer from a Zeiss fiber-optic based system which was taken off the coater. This unit was constructed using an integrating sphere, so that transmission and reflection can be measured simultaneously over a reasonable wavelength range of 300-1140nm. These data are then used to characterize the single layers.

A commercially available optical modeling package (FilmWizard[5]) was used to model both the single layers and the EC device. FilmWizard combines optimization and synthesis design capabilities with powerful analytical tools for interpreting spectroscopic and ellipsometric data, modeling of film layers, and performing regressions to determine actual thicknesses and indices of deposited layers. Each layer of the device was modeled successively and a model of the entire stack constructed. The results were relatively good, and compared well to the experimental measurements, so any improvements seen in the models are likely to result in real improvements in device performance provided realistic optical constants are used.

The model used employs approximately 8-10 layers in order to model each layer in the stack. The optical properties of the individual layers are first determined from the single film

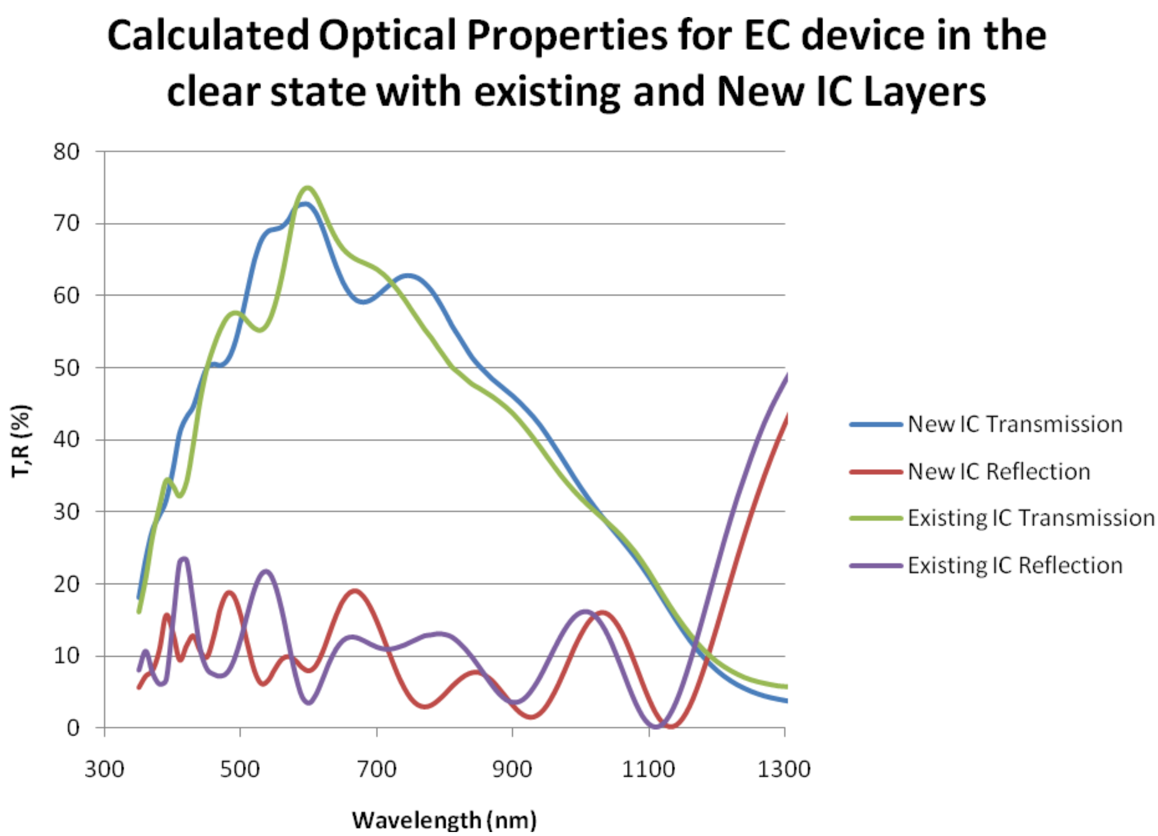

Figure 14 – Calculated optical behavior of an EC device in the clear state, modeled with existing and the novel IC material discussed in the text. It is clear that the transmission is slightly improved with the higher index material, while the amplitude of the reflection fringes are reduced, suggesting a less intense reflection color.

measurements, and then assembled into the complete model. Thicknesses of the layers are then adjusted to ensure that the model predicts the behavior of the overall stack as accurately as possible. Adjustments to the optical constants for the electrochromic layers are then made to predict the behavior in the colored state. A small selection of the results are shown in Figure 14 and Figure 15.

[5] FilmWizard is produced by SCI, http://www.sci-soft.com/Film%20Wizard.htm

It is clear from Figure 14 that there is a slight increase in the transmission for the device in the clear state, simply as a result of replacing the IC with a layer of higher refractive index compared with the existing IC material. Additionally, the amplitude of the fringes seen in the reflection spectrum indicate that the intensity of the reflection color will be lower, suggesting a more pleasing looking product.

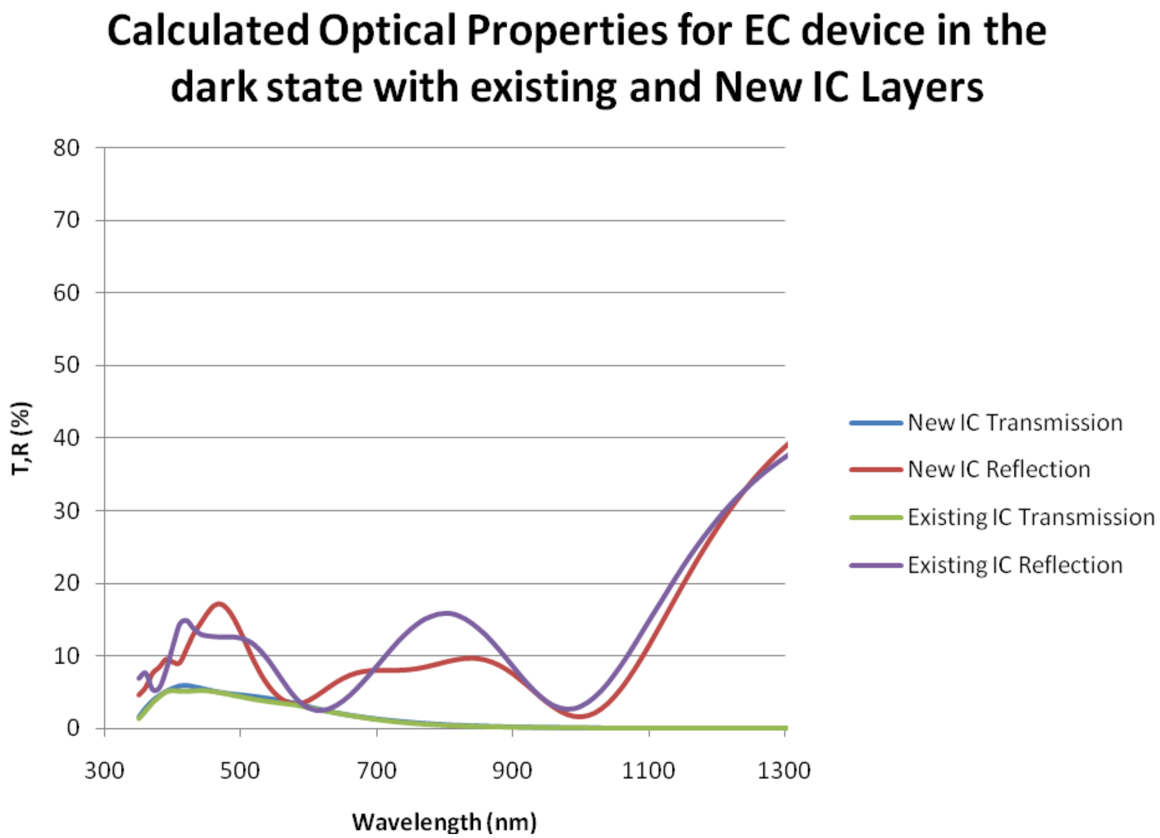

Figure 15 – Calculated optical behavior of an EC device in the dark state, modeled with existing and the novel IC material discussed in the text. It is clear that the transmission is largely unchanged with the higher index material, while the amplitude of the reflection fringes are slightly reduced, suggesting a less intense reflection color.

A calculation of the optical behavior in the colored state is shown in Figure 15 for a device with the existing IC and with the proposed new IC. It can be seen that the transmission is largely unaffected by the replacement of the IC, with the reflection fringes reduced slightly as a result of better matching between the refractive indices of the various layers.

It appears that the proposed changes to the IC material will have a beneficial effect on the optical properties of the device, in that the transmission in the clear state will be increased slightly and the reflection reduced slightly as a result of better optical matching between the various layers.

Task 3.3 Characterize Material Properties

The objective of this task is to utilize XPS (with ion milling), and dynamic SIMs at Evans Phi[6] to determine composition and structure of our film stack, and in particular the differences introduced as a result of the changes introduced due to the change in IC material.

We have used cross-sectional techniques for a number of applications, including analysis of shorts and defects, as well as for microstructural and microchemical analysis of different film stack designs (e.g. standard compared with sputtered IC) including both SEM (Scanning electron microscopy) and STEM (Scanning Transmission Electron Microscopy).

The data is particularly useful, in that we can gain both film thickness, microstructural information (grain size/morphology, porosity, surface roughness) as well as chemistry (atomic number) variations. A typical cross-sectional SEM image is shown in Figure 16.

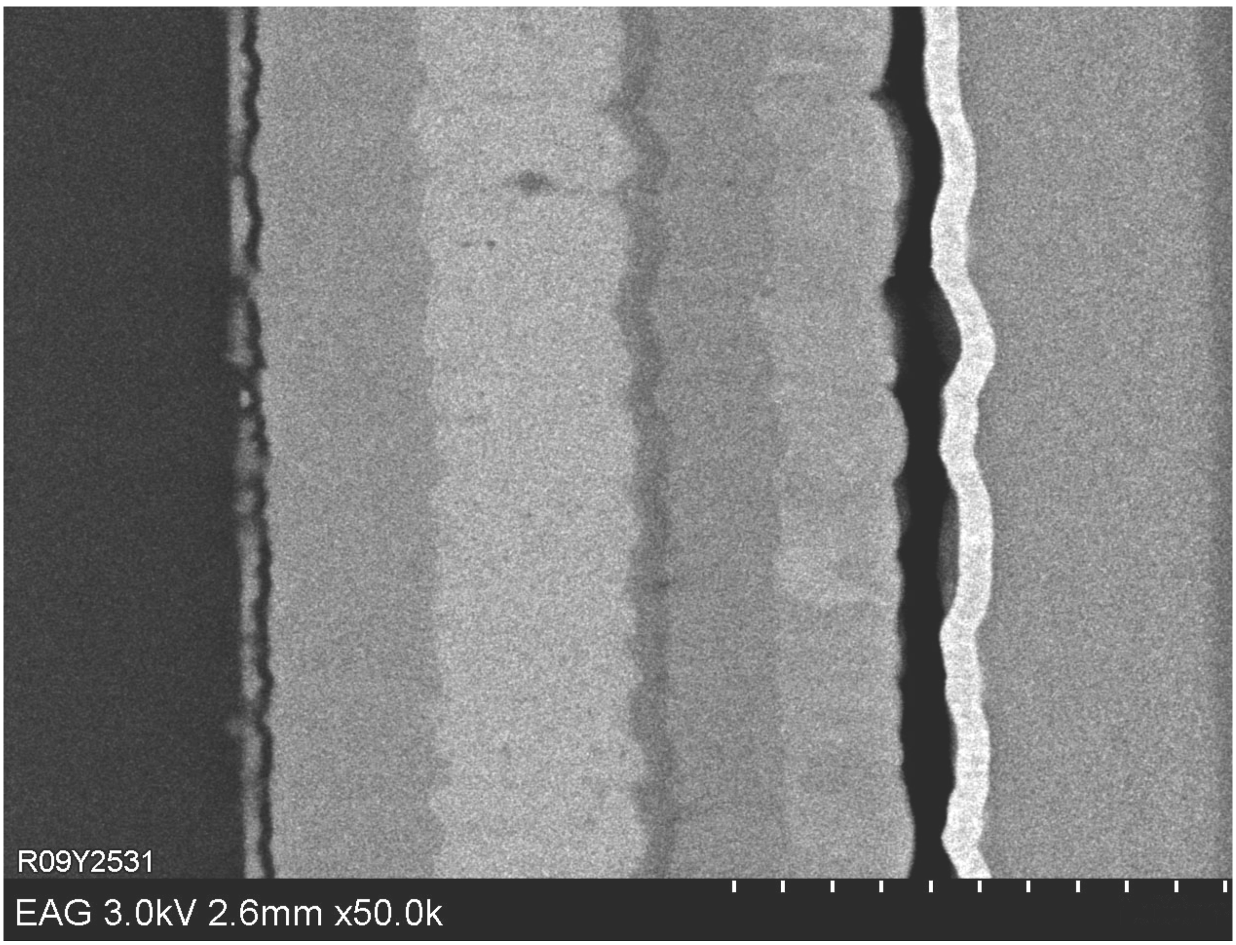

Figure 16 – Cross-sectional SEM image of the electrochromic device with sputtered IC layer described in the report. The substrate glass is the dark region to the extreme left of the image, and the films appear as bands of various shades of grey, the color of which is indicative of the average atomic mass of the layers.

When preparing samples for the transmission electron microscope (STEM), obtaining samples of uniform thinness (from 10–200 nm) is critical and time-consuming. There are a few different

[6] Fee for service analytical laboratory in Chanhassen, MN.

sample geometries, the most common of which is thin foils, or lamellae. A broad literature exists on the exact methods and some of the tradeoffs around creating lamellae. Mayer, Giannuzi, et al. (2007, MRS Bulletin) detail many of the more intricate decisions. The sample in Figure 17 was prepared using a focused ion beam (FIB). Opposing trenches are ion milled through the top of the sample using an ion beam (typically Ga or Ar), creating a thin electron-transparent sample. The advantages of the FIB technique include the speed of sample fabrication, large imaging areas, minimization of preferential sputtering and the ability to target a specific region for thinning to electron transparency (typically less than 100nm for most imaging modes). Several samples were prepared using this method.

Using one of these samples, we have measured the grain size in the TCO layers, porosity and compositional variation in the EC layer, thickness variation in the IC layer, apparent nanoparticles in the CE, and an interfacial zone between the CE and upper transparent conductive layer as is evident in Figure 17.

We have also used the approach to compare the microstructure of effect of TEC and our own transparent conductor on film interfacial roughness. In this case, we can see roughness is influenced by both the TEC and the EC layers. It has been determined that the roughness of the underlying films influence the roughness of the overall stack. In one example, a defect on the substrate surface that causes a hillock was seen to telegraph through the film, resulting in a hillock on the upper surface. This has significant implications for the expected haze level in the finished device, as we understand haze to be a strong function of surface roughness, and so having more control over the underlying substrate should enable better control of the finished surface roughness and resultant haze.

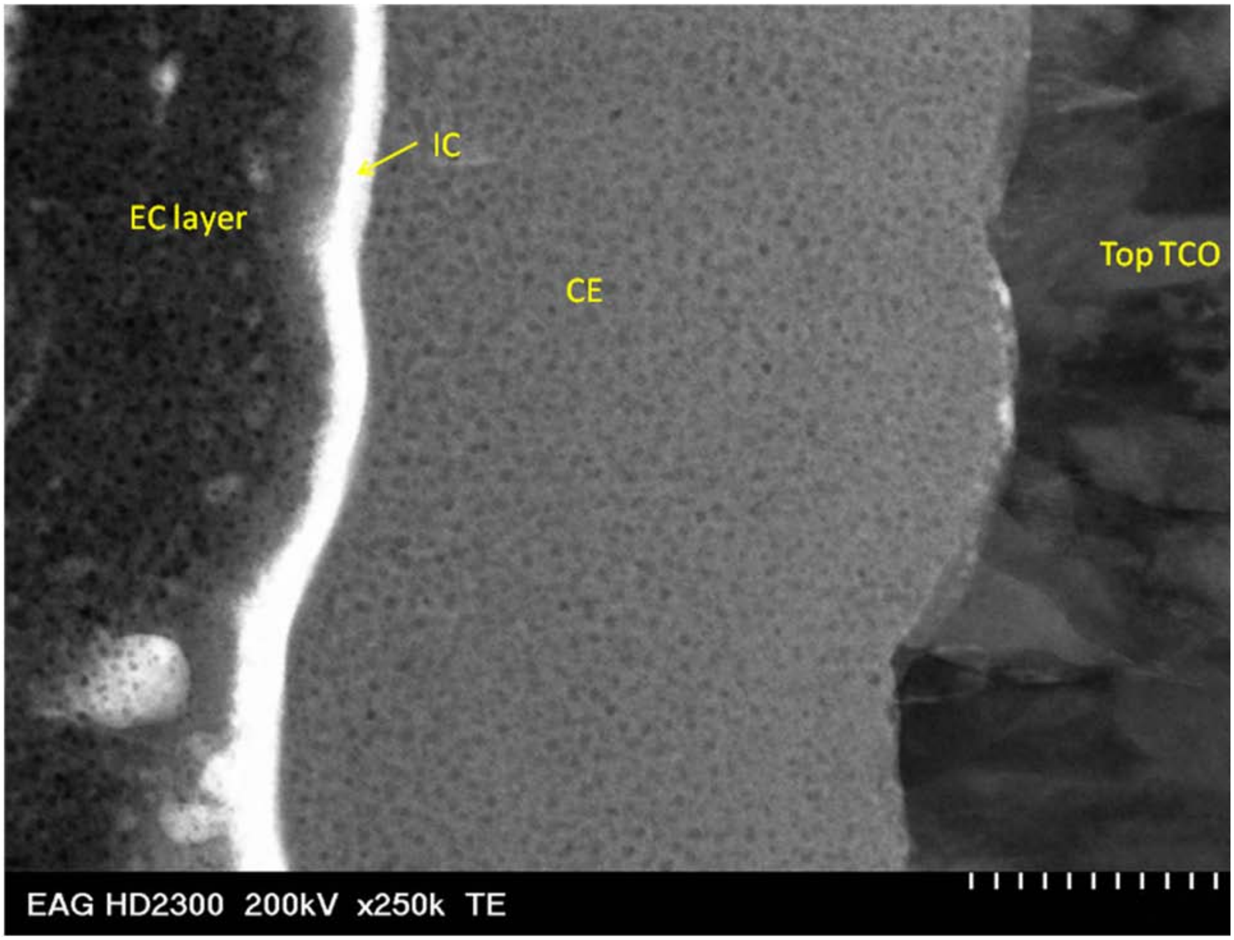

Figure 17 – High resolution STEM image of a part of the EC device focused on the CE layer, showing possible evidence of nanocomposite formation.

To summarize, a range of microanalytical techniques have been employed to characterize the structure and composition of the device stack yielding further understanding of key features of the films.

Task 4. Fabricate Devices into IGUs

This task is a matter of fabricating electrochromic devices into IGU's in order to allow some reliability tests to be carried out. Initially, the aim was to produce samples with dimensions around 10x12", but it was found convenient to increase the size to 18x18". The reasons for this are related to the availability of suitably sized carriers, and the fact that a target had been procured for the production coater, negating the need for the MRC lab coater to be used at all.

The reason for fabricating devices into an IGU is primarily to protect the films from exposure to potentially damaging environmental conditions, as well as to provide physical protection. SAGE has a well established technique to fabricate devices into IGU's, which involves applying busbars, protecting the device in a dessicated environment, making connections to the device by soldering wires to the busbars, and sealing the unit. Devices fabricated as part of this project went through this standard procedure with no significant modification to the process developed for the standard devices.

Task 4.1 Fabricate Small Area Samples

Initially, the task here was to fabricate 10 x12” samples based on the optimum film technology determined during the project, which was the largest size device that can be fabricated using the MRC coaters. It was found that deposition of films in the MRC was not required, so the size limitation no longer applied. Instead, a size suitable for production was chosen: 18x18”.

Several of these devices were fabricated, with the films deposited using a process developed as part of the previous task. The films were deposited entirely in the Production Coater, which although not configured optimally for this deposition sequence, allows us to produce acceptable quality films. These devices were then characterized in the monolithic state – that is before being fabricated into an IGU – using the in-house developed testing protocol which allows the electrical and optical behavior of the device to be determined.

The results were extremely encouraging, with some of the average performance numbers given in Table 1. Some of these devices have undergone cycling and re-characterization to establish some preliminary durability results. These are described in the following section.

Task 4.2 Carry out Room Temperature Cycling

Work at SAGE has included the investigation of the level of stress induced in a device for a given cycling protocol. Previous cycling results have quoted many thousands of cycles without significant changes in behavior. These cycles were carried out using a ‘short’ protocol, where devices are colored for approximately 7-10mins, and then bleached for the same time. It has been found that cycling devices in this manner does not cause them to reach full colorations, and so does not represent the most extreme stress that the device can be expected to experience.

A ‘long’ cycling protocol has been developed which requires the device be fully colored and held fully colored for a period of several hours. In the case of the work performed here, the samples are colored for 10 hours, and then bleached for 2 hours. This means that one ‘long’ cycle is equivalent in duration to approximately 50 ‘short’ cycles, although it is likely to be much more severe in terms of the stress induced in the films, and therefore more likely to lead to a stress induced failure than the short or shallow cycles.

The original target of 10,000 short cycles is therefore the equivalent of 200 ‘long’ cycles, which is still 100 days of two cycles per day.

Several devices (approximately 17x23” in size, two of which are shown in Figure 18) were fabricated with sputtered IC, according to the modified process developed above. These devices were characterized at the monolithic stage, and then fabricated. Following 31 deep cycles, the devices were retested. Essentially, the behavior of the devices remained largely unchanged, and if anything, some of the key performance parameters improved. One obvious change was for the initial transmission to be reduced by the addition of the matching lite as it has been made into an IGU. The differences were between 7.5 and 7.8%, which is as expected for a 92% transmitting matching lite. The switching times to both 5 and 10% were slightly faster after cycling, and the dark state transmission got lower after cycling – maybe as a result of the matching lite addition.

The colored state leakage current got slightly worse in two cases and better in one case, while the

Figure 18 – Some early 17”x23” samples produced as part of this work.

bleached state leakage current got slightly higher in two cases and better in one case. These changes are all relatively small, and are not thought to be indicative of a significant shift in the performance of the devices, particularly when the overall behavior is considered.

These devices went back on to deep cycling, and experienced 321 total cycles, with no significant impact on the performance. This is extremely encouraging, and strongly suggests that there is little or no impact on the durability of the EC devices produced with a sputtered IC layer when compared to the standard production method.

Task 5. Fabricate Large Sized Samples

In this task, large devices were fabricated using the pilot line production equipment. This followed readily from the work done in Task 3, however the size of the devices produced for this task approached the maximum size possible on the current equipment: 60” x 34”.

Task 5.1 Fabricate Large Glazings on Production Line

Figure 19 – Photographs of a large area device (60”x34”) produced using the techniques described in this report, shown in both the clear and colored states.

A large cathode of material C was installed into the production coater in a spare slot. This was used to produce large devices as shown in Figure 19. One of the devices is shown here in both the colored and clear states. In this case, the dimensions were 60” x 34”, which is approaching the maximum size for the present production equipment.

This particular device is characteristic of the majority of devices produced with sputtered IC in that it showed excellent uniformity, and was free of all of the banding defects which sometimes appear in the standard production devices.

Further runs were carried out to produce devices with a variety of different sizes, some of which have been subjected to durability testing, which is on-going, and there do not appear to be any significant difficulties with scale-up of the technology developed to date.

Task 5.2 Measure Defect Densities

SAGE has developed a digital camera system capable of measuring the size of defects, and recording their densities. The measured defect densities for the devices developed in this work compare very well with devices produced using the standard process, in that there are no significant increases in the observation. In fact, it is believed that at least one major class of defects will not be present in devices produced using the new technology as the defect formation mechanism has been clearly connected to one of the processes no longer required.

Task 5.3 Determine Optical Performance and Uniformity

The optical performance of the devices produced in this project does not show any significant differences from the standard production devices in terms of uniformity. The measurement of uniformity used in the glass industry is generally a measurement of ΔE, which is a calculated measurement derived from optical measurements. Transmission or Reflection measurements can be characterized by calculation of the CIE color coordinates: L*, a* and b*. These coordinates can be used to characterize any spectrum within the three dimensional space defined by L*, a* and b*. Any differences of color can therefore be calculated readily by using the following formula:

$$\Delta E = \sqrt{[(L_1^* - L_2^*)^2 + (a_1^* - a_2^*)^2 + (b_1^* - b_2^*)^2]}$$

where (L_1^*, a_1^*, b_1^*) represents one point in the color space, and (L_2^*, a_2^*, b_2^*) the other.

Choice of two extreme points in the color space will yield the maximum value of ΔE, and this can be used as a measurement of uniformity. There are some issues with this measurement due to the fact that abrupt changes of uniformity can have the same value of E as more gradual changes, which may or may not be seen differently in terms of visual acceptability. However, for the purposes of this exercise, we use a measurement of E to compare between devices produced using standard technology and the devices developed as part of this project.

Several devices have been produced, and the optical properties measured. There is no apparent difference between the uniformity measured in both the colored and clear states between the new and old technology. As an example, a device which was 42” x 27” had a measured ΔE of 7.3 in the clear state after one color-bleach cycle. This should be compared with an average of 4±2 for standard production. The small increase for the norm is probably due to the influence of a few minor defects which can easily be reworked. This is shown, because the next measurement after 57 deep cycles, gave a value of ΔE=3.4.

As we have seen in previous sections, the optical performance of the devices produced with the novel IC layer show very similar behavior to those produced using the standard method. There is one exception to this, where the reflection is slightly lower in the clear state as a result of the better matching of optical constants within the electrochromic stack, which is an improvement on the existing product performance.

Task 5.4 Durability Testing

The aim of this task was to carry out durability testing, which is based predominantly on room temperature cycling. Several devices were produced and subjected to color-bleach cycling and the results described here.

The devices (which were 18x35") were fabricated into IGUs, had the defects reworked, and have been cycling since 12 December 2006. They were initially characterized using our standard device characterization. They were then cycled using the following protocol: +3V applied voltage for 7mins, followed by –2V for 7 minutes. The devices were subsequently re-characterized after 184, 279, and 350 cycles. A further device continued cycling, and was characterized after approximately 22000 cycles, which exceeds the number actually required for successful completion of the task.

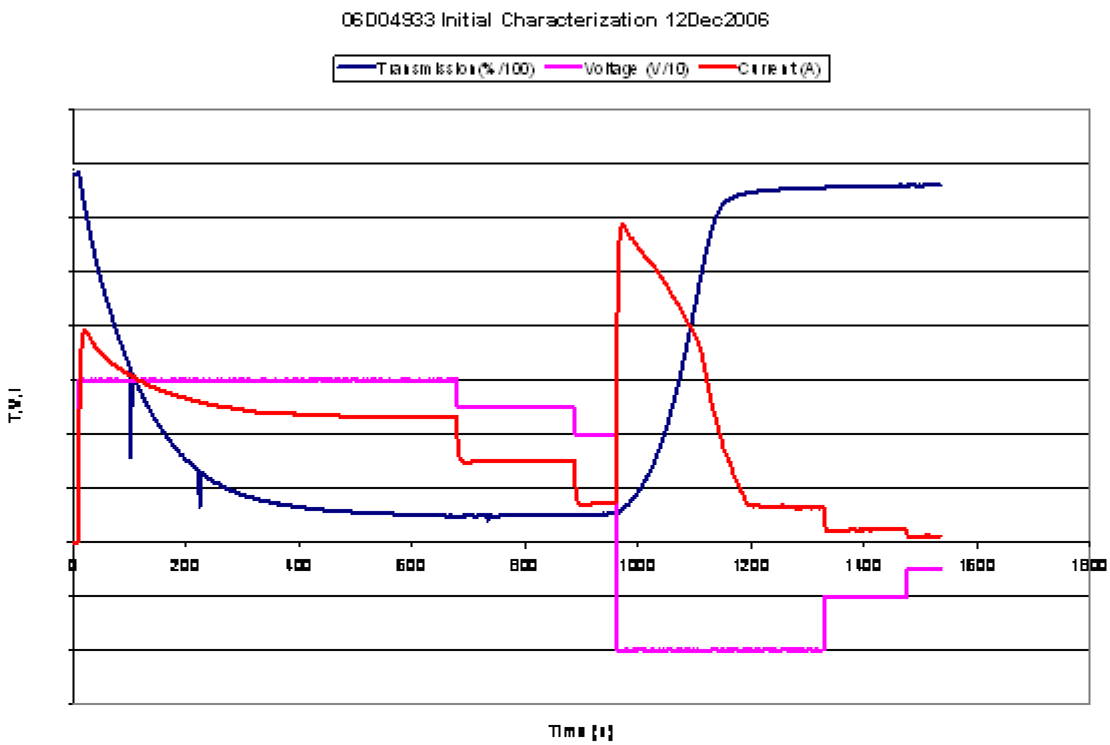

Figure 20 – Initial characterization measurements for a device produced using the novel technology, taken prior to subjecting the device to room temperature cycling. Current is shown in red, voltage in pink and transmission in blue.

In the initial characterization test, the device is first colored with 3V, then the voltage is stepped down to 2.5V and the 'equilibrium' current and transmission measured. Then it is stepped down again to 2V, and the equilibrium measurement repeated. Next, the device is bleached with -2V, and again the voltage stepped down to –1V and then –0.5V. Transmission and current are simultaneously measured. In this figure, the current is shown as positive for both coloring and bleaching, but is in fact negative for the bleaching phase.

Some typical results are shown in Figure 20. The current is shown in red, and shows typical electrochemical characteristics, with increases in current as a response to a step change in voltage, followed by an equilibration period, and then a stable region. The transmission response is as expected, with the transmission decreasing as the integral of the charge passed through the device.

The characterization after 350 cycles is shown in Figure 21, this time for two devices. It can be seen that very little has changed, both in terms of the dynamic range and the switching speed, and that the repeatability between devices is also very good.

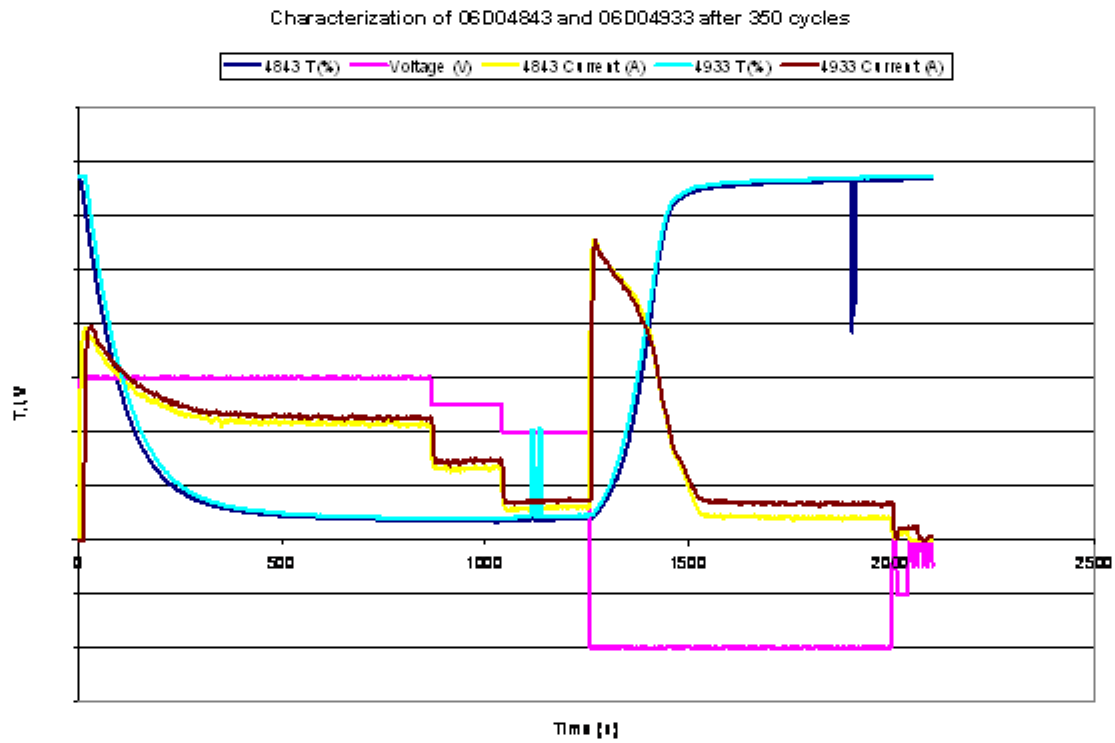

Figure 21 – Characterization data for devices after 350 short color-bleach cycles, showing remarkable consistency between devices, and also little change for the data shown in Figure 20.

In order to demonstrate this more clearly, the current for the device shown in Figure 20 before and after cycling is plotted in Figure 22. The current and the transmission are plotted for the tests shown in the previous two figures, and can be seen to be replicate one another very closely, indicating the stability of the device to cycling. In fact, the colored state transmission can be seen to improve (become lower) very slightly as a result of cycling, and the leakage current drops slightly, which is also an improvement.

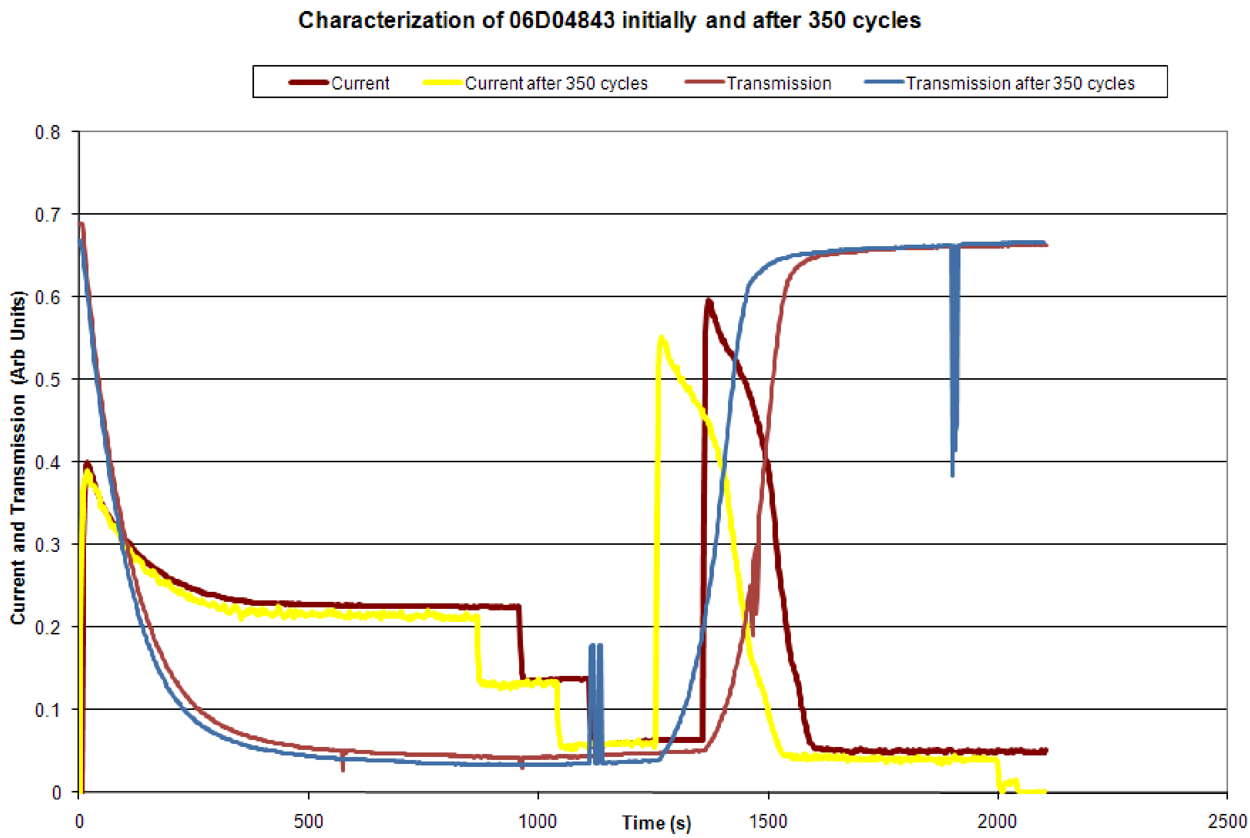

Figure 22 – Comparison of the behavior of one of the devices discussed in the text, showing the current and transmission at the start of the test and after 350 short cycles. There is little significant difference between the plots, and if anything, the dynamic range and leakage current can be seen to have improved slightly with cycling.

As mentioned above, further cycling measurements were carried out on a device for up to 22000+ cycles. This device showed excellent stability after this period, as illustrated in Figure 23, which shows key measurements taken during the duration of the test.

This device shows excellent durability and stability over the 22000 cycles of the test, and there is no reason to suspect that this should not hold for further cycles. In particular, it can be seen that the critical performance parameters – the colored and bleached transmission, coloration time and the leakage currents – all stay remarkably constant and within the measurement tolerance of the experiment. In fact, several of these measurements indicate an improvement in the device performance as cycling proceeds. The only parameter which appears to be getting worse is the dark state current, but the value at 22000 cycles is still lower than that at around 5000 cycles, and the change is actually quite small relative to the magnitude of the current being measured.

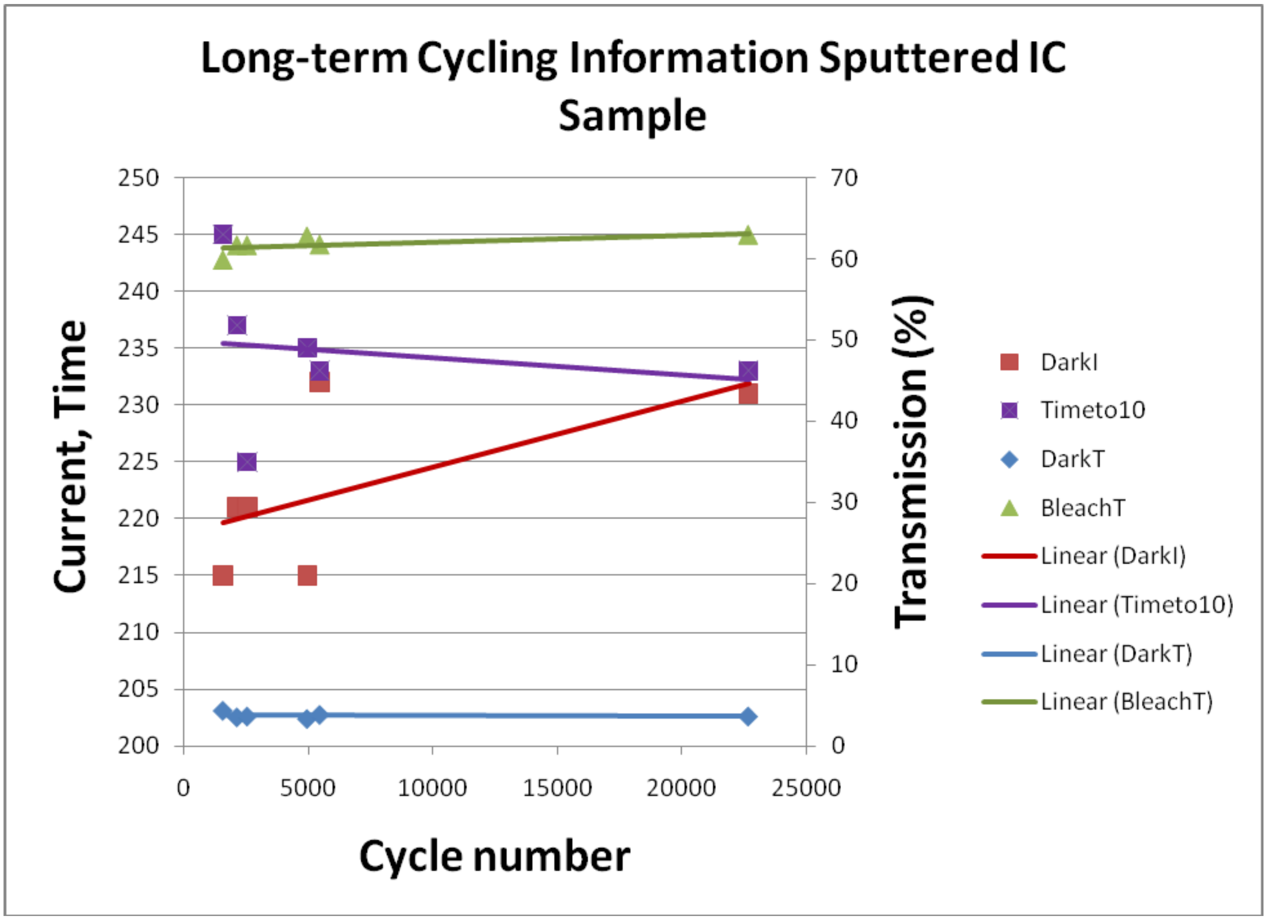

Figure 23 – Behavior of the critical characteristics for a sputtered IC device subjected to prolonged long term cycling up to around 25000 cycles. It can be seen that the critical parameters of transmission and Leakage current, which have been the characteristics which reflect any degradation are largely unchanged during this period.

Originally, the task called for 10,000 short cycles to be completed to fulfill the task. Figure 23 shows that this has been completed satisfactorily, and in fact the cycling progressed to 22000 cycles with no apparent degradation.

In order for this to be completed in a reasonable length of time, the cycles are consequently short – of the order of 15 minutes – and this means that the devices do not necessarily color or bleach fully. Although the success criterion of over 10000 short cycles has been achieved, it was felt that this was not an adequate durability test as it did not replicate the cycling protocol which was likely to be seen in the field. In fact, several devices developed shorts when exposed to additional cycling after the 'so-called' burn-in test, which was supposed to develop them all to allow rework prior to shipment.

Subsequently, a separate study within SAGE has determined that this short cycling protocol is not the most effective way of screening for infant mortality defects, and so the cycling test has been universally replaced with a test with a longer cycle time – of around 10 hours per cycle – which ensures the device fully colors and also more closely represents the typical end user cycling protocol.

In the test which led to adoption of this change, carried out in late 2007, the objective was to measure the rate of short generation for the devices which were considered to be 'Product Quality' at that time. Approximately 90 devices were used which had already undergone the 'short cycle' burn-in protocol of between 700 and 1000 (15 minute) cycles. The test consisted of inspecting the devices after a certain number of cycles, and counting the number of shorts each had developed. Each short was then marked so that it would not be recounted, so the measurement was of shorts generated during the preceding period. An example of the data is shown in Figure 24 where the rate of short generation per unit area per number of cycles is plotted against number of cycles. This plot shows the rate on a log scale.

The important point to note, however, is that there are a significant number of shorting defects which appeared AFTER the (then current) short cycle burn-in period of 700 to 1000 cycles. For example the value of 0.138 shorts per square foot per cycle represents about 1 short per device per cycle. This is an enormous number which did not appear during the accepted burn-in procedure, and it led to the adoption of the present longer cycle times for the burn-in, as well as a significant change in the device fabrication process, which is outside the scope of this report, but was intended to reduce the overall level of contamination which is directly responsible for these types of defects.

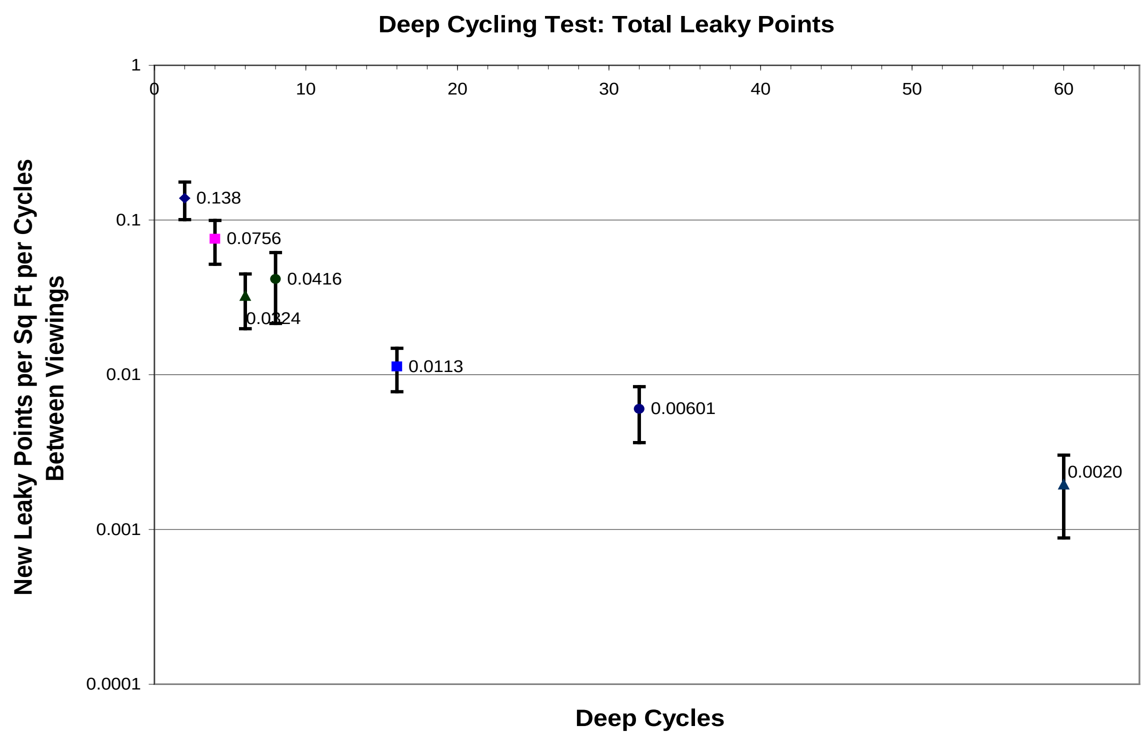

Figure 24 – Total number of leaky points developed during 'long' cycling AFTER the then standard burn-in procedure had been completed. This shows that an enormous number of latent shorts were still present in the devices after the burn-in procedure had been completed, which can lead to significant numbers of failures in the field.

As a result of this change, all the subsequent reliability cycling data are taken using the longer cycling procedure. Current data are taken on devices which are being generated from time to time as a result of Process optimization DOE experiments, which yield devices with reasonably representative make-ups and process parameters.

To date, no significant issues with the new process have been detected. In particular, the physical integrity of the IGU of the thin films has not been compromised as a result of the changes outlined in this report. This has included investigations of the performance as described above, the bus bar adhesion, and the IGU integrity. From these measurements, it appears that there are no significant issues with durability of the new technology, and it is anticipated that further reliability tests will serve to confirm this.

Discussion and Conclusion

This project has enabled development of the next generation of electrochromic devices suitable for large-scale production. Specifically, the requirements to produce large area devices cost effectively require processes amenable to mass production, using a variety of different substrate materials, having minimal handling and capable of being run at high yield. The present SageGlass® production process consists of two vacuum steps separated by an atmospheric process. This means that the glass goes through several additional handling steps, including venting and pumping down to go from vacuum to atmosphere and back, which can only serve to introduce additional defects associated with such processes. The aim of this project therefore was to develop a process which would eliminate the need for the atmospheric process.

The overall project was divided into several logical tasks which would result in a process ready to be implemented in the present SAGE facility. Tasks 1 and 2 were devoted to development and the optimization of the IC material process. These tasks are more complicated than would be expected, as it has been determined in the past that there are a number of interactions between the IC and the layers beneath, which have an important effect on the behavior of the device. The effects of these interactions needed to be understood in order for this task to be successful. Tasks 3 and 4 were devoted to production of devices using the novel technology developed in the previous tasks. In addition, characterization tests were required to ensure the devices would perform adequately as replacements for the existing technology.

Each of these tasks has been achieved successfully. In task 1, a series of potential materials were surveyed, and ranked in order of desirability. Prototype device structures were produced and characterized using a variety of techniques, including sophisticated high resolution microscopy techniques as well as standard electrochemical, optical and electrical measurements, in order to do this. These diverse activities satisfied the requirements for Task 1.

From the results of this relatively extensive survey, the number of candidate materials was reduced to one or two. Small devices were made in order to test the functionality of such samples, and a series of optimization experiments were carried out with encouraging results. Devices were fabricated, and some room temperature cycling carried out showing that there are no fundamental problems with this technology. This series of achievements satisfied the requirements for Tasks 2 and 3.

The results obtained from Task 2 naturally led to scale-up of the process, so a large cathode was obtained and installed in a spare slot in the production coater, and a series of large devices fabricated. In particular, devices with dimensions of 60” × 34” were produced, using processes which are fully compatible with mass production. Testing followed, satisfying the requirements for Task 4.

Table 2 shows the proposed project success criteria for Task 4, along with the achieved performance from a representative device. These numbers were initially proposed for a 10”x12” device, and the majority of the work has been carried out on a slightly larger size of 18”x18”. It would therefore be anticipated that some of the parameters would scale by a factor of about 2.7 which is the ratio of the areas.

Parameter	Target	Task 4 Performance	Comment
Threshold Voltage (V)	2.0V	2.0V	
Optical Density	>1.4	1.48	$Log_{10}(T_{ble}/T_{col})$
Coloration time (s)	120	188	Slightly longer due to larger size of 18x18”.
Leakage Current at 3V (mA)	~50mA	61mA	Slightly higher due to larger device size
Bleached Transmission (%)	>70%	72%	
Uncorrectable defects after 10000 cycles	0	0	

Table 2 – Collected data for devices produced as a result of work carried out in Task 4. Note all these parameters were measured on a single device. Other devices have shown improvements in each of the areas, but these data are given as generally representative of the current state of the process.

As can be seen from the table, each of the proposed success criteria have been met or exceeded. It should be noted that all these measurements were taken on the same device. Other devices have shown improvements in each of the areas, but these data are given as generally representative of the current state of the process. It is clear that the success criteria have largely been achieved, and it could be argued that the larger sized device could be responsible for the leakage current and switching time being slightly outside the required levels.

Further reliability testing will be required, but to date there has not been anything to suggest that there are any significant differences expected between this novel technology and the existing excellent durability results for the current production technology.

As can be seen from this discussion, all the requirements of the project have therefore been successfully achieved. The devices produced using the newly developed technology showed excellent optical properties, often exceeding the performance of the existing technology, equivalent durability results, and promise a significantly simplified manufacturing approach, thereby suggesting higher yields as a result of less handling, and therefore lower costs.