

SIZO/Ag/SIZO/NiO sandwich film based smart window combining hybrid low emissivity and electrochromism for the dynamic control of solar radiation

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ARTICLE INFO

Handling Editor: Dr P. Vincenzini

Keywords:

Nickel oxide
Electrochromic devices
Smart window
Sputtering
Low emissivity

ABSTRACT

Low-emissivity (Low-e) smart windows are a promising option for effective thermal regulation, aimed at reducing energy consumption and enhancing interior comfort via their ability to modulate sunlight. To improve the sunlight modulation (VIS) and thermal management (NIR) of smart windows, we present an assembly of amorphous SIZO/Ag/SIZO/NiO electrode to develop Low-e electrochromic windows for the dynamic control of solar radiation, fabricated via sputtering technology at room temperature. The fabricated device demonstrates an exceptionally high visible modulation (78 %), providing clear visibility in windows, and superior near-infrared (NIR) shielding efficacy (>80 % in whole scale), good cyclic stability, with an ultrafast switching speed of 1 s for bleaching and 3 s for coloration. The capacitive electrochromic electrode, with a specific capacitance of 220 Fg⁻¹, exhibits enhanced electrochemical kinetics, substantial charge capacity, and is suitable for application as a secondary energy storage device. The beneficial, extremely disordered amorphous structure, coupled with an enormous quantity of defects, promotes ultra-fast ion intercalation and de-intercalation processes with enhanced electrochromic efficiency. This research signifies a notable progression in the fabrication of Low-e electrochromic films, which will illuminate advancements in electrochromic technology for future energy-efficient smart buildings.

1. Introduction

In today's modern world, the issue of energy concerns has undeniably emerged as a significant concern, garnering global attention. Buildings are responsible for about 40 % of worldwide total consumption of energy and 26 % of global energy-related emissions [1–3]. This includes both direct emissions from buildings themselves (accounting for 8 % of total emissions) and indirect emissions resulting from the generation of electricity and heat utilized in buildings (accounting for 18 % of total emissions). The energy consumption in the buildings industry had a modest rise of around 1 % in the year 2022 [4]. Specifically, relying too much on air conditioner to keep rooms at a comfortable temperature uses a lot of electricity and releases a lot of greenhouse gases [5]. Although buildings use about 40 % of the world's energy, windows, which are one of the least energy-efficient parts are responsible for the 60 % of energy loss [1–3]. It is estimated that houses in the United States use about 4 % of the country's total energy for heating and

cooling through windows [6]. Hence this decade is very important for taking the steps needed to build buildings that use less energy and make 20 % of current buildings energy efficient by 2030. In this regard, for global decarbonization and energy saving it is imperative to develop energy efficient smart windows [7]. Research carried out in this area have demonstrated that installing smart windows with reversible optical qualities can lead to a 50 % decrease in the amount of energy used in buildings for lighting, heating, and cooling [8–11]. Switchable smart glass functions by enhancing energy efficiency by the selective obstruction of heat and light whenever needed, so mitigating the excessive utilization of heating, air conditioning, and lighting systems. The utilization of the completely opaque privacy mode results in a 40 % reduction in heat penetration into the building's interior. Daylighting in buildings mostly depends on the visible light spectrum of solar radiation, which ranges from 400 to 760 nm. The remaining 50 % of solar radiation is predominantly found in the near-infrared range (NIR, 760–2500 nm) and is mainly used for indoor heating purposes [12,13]. The ability of

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<https://doi.org/10.1016/j.ceramint.2025.04.059>

Received 19 December 2024; Received in revised form 24 March 2025; Accepted 6 April 2025

Available online 7 April 2025

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controlling the transmission of near-infrared (NIR) light via windows has a substantial impact on the regulation of interior temperatures and the conservation of energy. It is important to mention that a little adjustment of 1 °C in interior temperature can lead to a significant decrease of 10 % in energy consumption [14,15]. Low-emissivity glass, commonly referred to as Low-E glass, are such type of energy-efficient glass which can effectively obstruct around 90 % of solar radiation, therefore maintaining inside heat during cold weather and preventing heat from entering during hot weather. Smart windows based on electrochromic glass along with low-emissive property are a promising technique for reducing buildings' energy usage [16].

Electrochromic devices (ECDs) can alter their optical characteristics (such as absorbance and transmittance) in response to an external voltage [16]. Among the broad category of chromogenics, ECDs commonly exhibit a 5-layer structure that bears a striking resemblance to that of an electrochemical battery [17]. Electrochromic (EC) devices typically consist of two transparent substrates that are rendered conductive through the deposition of thin films of conductive transparent oxides, such as fluorine-doped tin oxide (FTO) or indium tin oxide (ITO). These substrates are then layered with two materials that exhibit complementary electrochromic properties, namely tungsten trioxide (WO₃) as the anodic material along with nickel oxide (NiO) as the cathodic material [18].

Nowadays, the predominant transparent electrode utilized in EC devices is indium-tin-oxide (ITO) owing to its favorable characteristics including low resistivity, high transparency, and well-established mass manufacturing techniques like sputtering [19,20]. Although amorphous ITO (a-ITO) films may be efficiently deposited using traditional DC sputtering at room temperature, thick a-ITO films are not suitable for transparent and flexible ECDs because of their brittleness and high sheet resistance [21–23]. When some of semiconducting oxides like IZO or AZO (Indium or Aluminium doped Zinc Oxide) are heavily doped, they forfeit their semiconducting capabilities and transform into "degenerate-semiconductors" with excellent conductivity, like that of metallic materials [24]. Only a small number of materials fall into this category of transparent conductors, as typically more doping leads to enhanced conductivity but decreases optical transmission, and vice versa. Therefore, it is challenging to get both high electrical conductivity and high optical transparency in the visible spectrum.

As a replacement for a-ITO films, Metal oxide/metal/metal oxide (OMO) multilayer structures are extensively studied as high performance transparent conductive electrode (TCE) and in optoelectronic devices [25–29]. Some major advantages of OMO multilayer structured electrodes include high transparency in visible region, low resistivity, enhanced mechanical property, lower overall thickness, and applicability for ordinary temperature processing, etc. [30–32]. Several investigations have been conducted on the fabrication of transparent EC devices utilizing OMO multilayer electrodes [33,34]. However, there is a significant demand for the fabrication of conducting layer with high transparency, high electrical conductivity, and with NIR modulation or Low-emissivity (Low-e) property for energy efficient ECDs. Low-e coatings were specifically developed to mitigate heat losses, hence enhancing the heat insulating capabilities of window glass [35]. For example, radiative heat emitted by the external surroundings may be efficiently insulated to maintain a cool temperature in the interior throughout the summer. Under winter conditions, it is necessary to decrease heat conduction via a narrow window to maintain a warm and stable interior temperature [36].

This work introduces a multilayer design consisting of oxide-metal-oxide (OMO), SIZO/Ag/SIZO (SAS), where SIZO (Si–In–Zn–O) indicates the incorporation of Si into an In–Zn–O system composed of SiO₂:In₂O₃:ZnO = 1: 74.5: 24.5 at wt%) layers as a transparent conducting oxide (TCO) material. This material exhibits amorphous nature with Low-e property and boasts superior electrical, and optical properties. The objective of this research endeavour is to fabricate energy efficient Low-e electrochromic devices. We demonstrate the

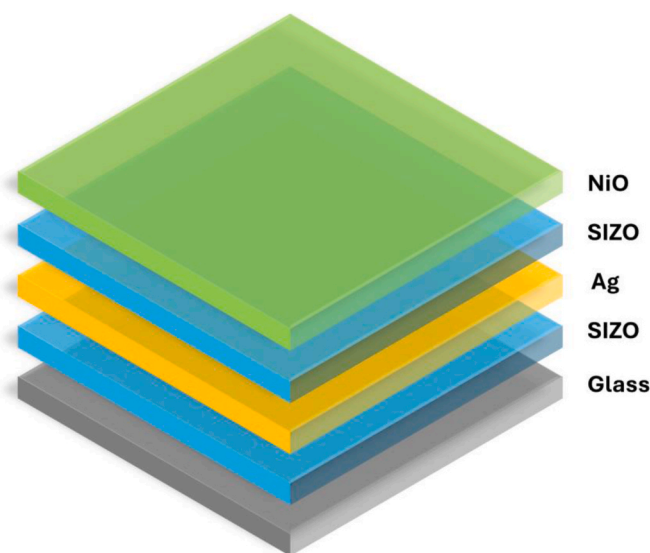


Fig. 1. Schematic illustration of electrochromic Low-e SIZO/Ag/SIZO/NiO electrode structure.

electrochromic characteristics of nickel oxide (NiO) thin films deposited onto glass substrate coated with SAS multilayers. The deposition process was carried out utilizing an ambient temperature RF sputtering system. The fabricated film demonstrated superior electrochromic performance, achieving optical modulation of 78 % at 550 nm (VIS), 44 % at 950 nm (NIR), and 3 % at 2400 nm (NIR), along with commendable cycle stability. The designed electrochromic low-emissivity films for building wall thermal envelopes provide a solution for year-round energy savings in heating and cooling.

2. Experimental methods

2.1. Preparation of SIZO/Ag/SIZO multilayer film

An ingenious SIZO (Si–In–Zn–O) ceramic target, measuring 2 inches in diameter and 5 mm in thickness, was designed for regulating conductivity and transparency. Silicon was incorporated into an In–Zn–O system (SiO₂:In₂O₃:ZnO = 1:74.5:24.5 at wt%). The SIZO OMO multilayer thin films were successfully fabricated onto a glass substrate using multi-sputter (DaeKi Hi-Tech). The deposition of an amorphous oxide layer was carried out using RF magnetron sputtering using Si–In–Zn–O target, whereas the Ag layer was deposited by direct current (DC) magnetron sputtering using Ag (99.99 % pure) metal target (2" diameter and 5 mm thickness) under room temperature. SIZO layer deposition were carried out for 35 min under a working pressure of 6×10^{-3} Torr, RF power of 40 W. The DC sputtering parameters used for silver (Ag) deposition were as follows: a base pressure of 5×10^{-6} Torr, a sputtering power of 20 W, a working pressure of 4×10^{-3} Torr. Both SIZO and Ag layer deposition were carried out under pure argon atmosphere.

2.2. Preparation of NiO thin films

Thin films of nickel oxide (NiO) were deposited via RF magnetron sputtering with a 99.9 % pure nickel oxide target (2" diameter and 5 mm thickness). Glass precoated with SIZO/Ag/SIZO (sheet resistance 4.81 Ω/cm^2) was used as substrate. The target-substrate distance was 17 cm, and all the substrates were placed in the vacuum chamber evacuated to 5×10^{-6} Torr without additional heating. Then inert Argon gas was pumped into the chamber. The deposition pressure was 16 mTorr and the gun power for the NiO target was 60 W. The deposition was carried out for 120 min. The schematic illustration of multi-layer structure of

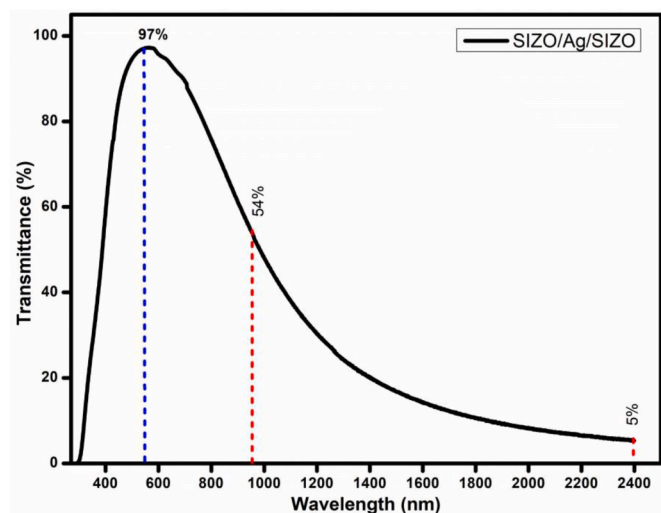


Fig. 2. Transmittance spectra of Low-e SIZO/Ag/SIZO substrate.

SAS-NiO thin film is shown in Fig. 1.

2.3. Measurements

The structural property of NiO thin film was studied by X-ray diffraction (XRD). Investigation of visible and IR transmittance of OMO and EC layer was conducted using a UV–Vis optical spectrometer (Cary 5000 UV–Vis–NIR, Agilent) in the wavelength range of 300–2400 nm, while electrical parameters for OMO were examined using Hall measurement (HMS 3000, Ecopia). The SIZO OMO structure was precisely validated by transmission electron microscopy (TEM, Titan cubed G2 60–300, FEI business). The surface morphological property of the film was studied by Field emission scanning electron microscopy (FESEM) equipped with energy dispersive X-ray analysis (EDX) (FEI QUANTA 250, Czech Republic). The elemental composition of the deposited film was studied by X-ray photoelectron spectroscopy (XPS: JEOL PS-9010 TR). The cyclic voltammetry (CV) of the film were studied using EC system SP-300 potentiostat (Biologic Science Instruments, France) in conjunction with EC lab software. Chronocoulometry (CC) and chronoamperometry (CA) were performed using an electrochemical workstation scanning potentiostat/galvanostat (CH Instruments Inc., USA; Model: CHI604D). Coloration and bleaching of devices were achieved by subjecting them to external voltages of +3 V and −3 V, respectively, using an DC source.

3. Results and discussion

3.1. Optical and electrical analyses of the SIZO/Ag/SIZO multiple layers

SIZO/Ag/SIZO multilayer structures include all the necessary characteristics, and, despite the higher number of layers, the overall electrode thickness is decreased. If the typical thickness of the individual films of ITO, FTO, and AZO falls between 150 nm and 250 nm, our SIZO/Ag/SIZO multilayer structures consist of just 46.9 ± 2.3 nm thick oxide layers combined with a 10.7 nm metallic interlayer. Consequently, the overall thickness of the SAS multilayer electrode is about 104.5 nm. Hence, when comparing the amount of ITO required for a single layer (approximately 150–160 nm) with the amount of SIZO utilized in a multilayer electrode consisting of SIZO/Ag/SIZO (104.5 nm), it becomes evident that the quantity needed for the fabrication of the multilayer electrodes is reduced by a factor of 2. The electrical properties of SAS multilayer showed sheet resistance of $4 \Omega/\text{sq.}$, which is much lower than the commercial ITO [37], bulk concentration of $-1.71 \times 10^{+22} \text{ cm}^{-3}$, conductivity of $2.57 \times 10^{+4}$, and mobility of $9.39 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

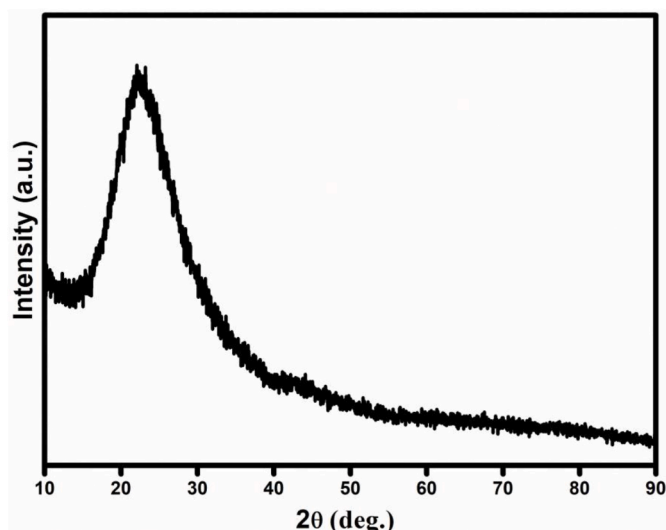


Fig. 3. X-ray diffraction pattern for NiO thin film deposited on glass substrate.

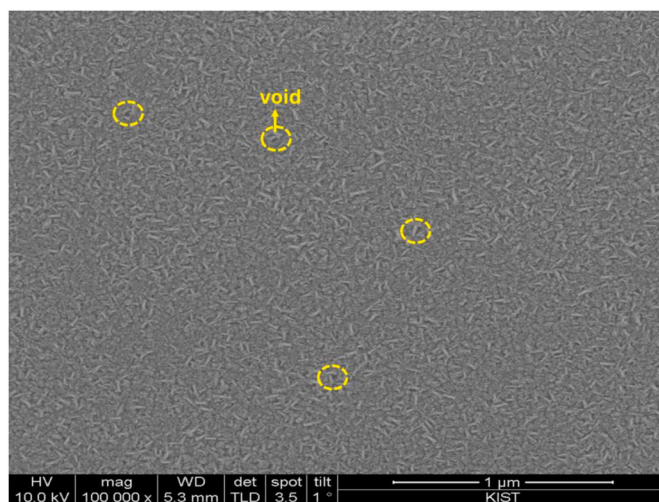


Fig. 4 (a). FESEM image of SIZO/Ag/SIZO/NiO thin film.

As reported in our previous study [38], XRD patterns of SIZO/Ag/SIZO multilayer films showed amorphous nature. Fig. 2 displays the optical transmittance spectra for SIZO/Ag/SIZO multilayers across a wavelength range of 300–2400 nm. A low-emissivity coating is a protective layer applied to window glass that is transparent in visible light (380 nm–780 nm) and strongly reflects light in the infrared (IR) range. Its purpose is to minimise heat loss in the form of radiant heat [39]. Fig. 2 illustrates that the SAS film exhibits high transparency in the visible spectrum and significantly reduced transmittance in the near-infrared (NIR) range. Specifically, it achieves a transmittance of over 97 % at 550 nm, 54 % at 950 nm, and progressively drops to 5 % at 2400 nm.

3.2. Structural and morphological analysis

The electrochromic properties of NiO films are significantly influenced by their crystal structure and chemical bonding. Fig. 3 shows the X-ray diffraction pattern for NiO thin film deposited on glass substrate indicating the amorphous nature. Amorphous thin films, due to the disordered configuration in the structure can, facilitate numerous open voids for ion transport, enable rapid ion migration, and offer low energy barriers for ion insertion and extraction [40]. This configuration reduces

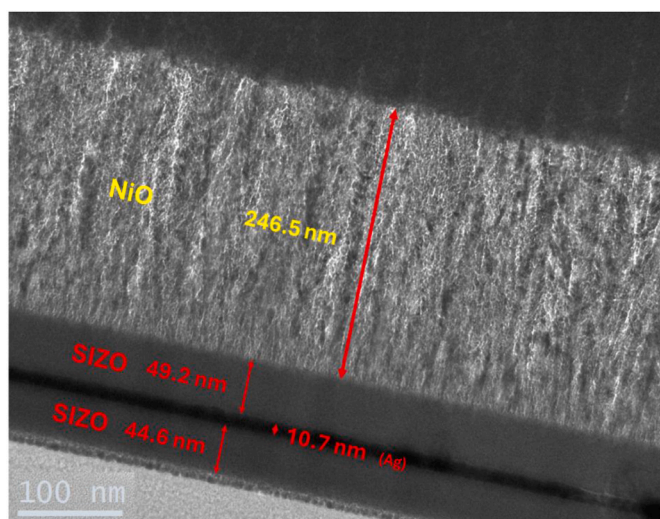


Fig. 4 (b). TEM image of SIZO/Ag/SIZO/NiO thin film.

the diffusion path length of ions and enhances the active specific surface area for reactions, resulting in superior colouring efficiency and exceptional electrochromic properties [41]. Arakaki et al. observed that the amorphous NiO films exhibited enhanced electrochromic efficiency when compared to cubic crystalline NiO film cycled in a 0.1 M KOH electrolyte [42]. Therefore, the developed amorphous NiO thin film may facilitate the rapid flow of electrolyte ions inside the film, allowing for a higher number of ions to be accommodated and enhanced optical modulation [43,44]. Fig. 4 (a) illustrates the surface morphology of the nickel oxide film. This image indicates that the films have considerable uniformity, including surfaces like triangular pyramids with voids that permit efficient ion intercalation, hence enhancing electrochemical efficiency.

The structure and thickness of SIZO OMO, NiO was evaluated using a cross-sectional TEM picture seen in Fig. 4 (b). The overall thickness of SIZO OMO was found to be 104.5 nm, with approximately 11 nm Ag layer. The thickness of NiO is found to be 246.5 nm. Fig. 4 (b) shows a well-defined layered structure of SIZO/Ag/SIZO/NiO with a distinct interface between each layer.

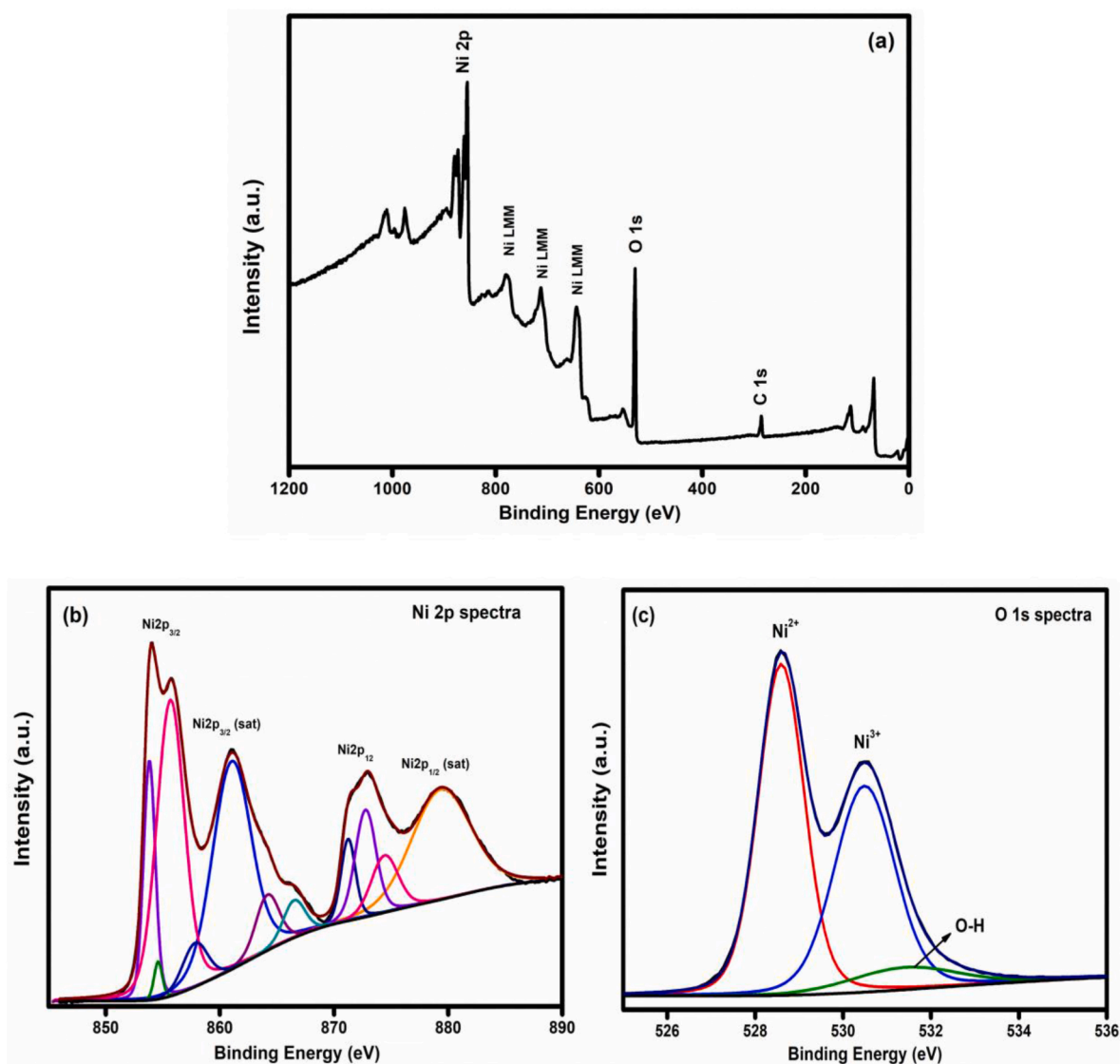


Fig. 5. (a) XPS survey scan spectrum of NiO thin film, (b) XPS core-level spectrum of Ni 2p states in NiO thin film, and (c) O-1s core-level spectrum of NiO film.

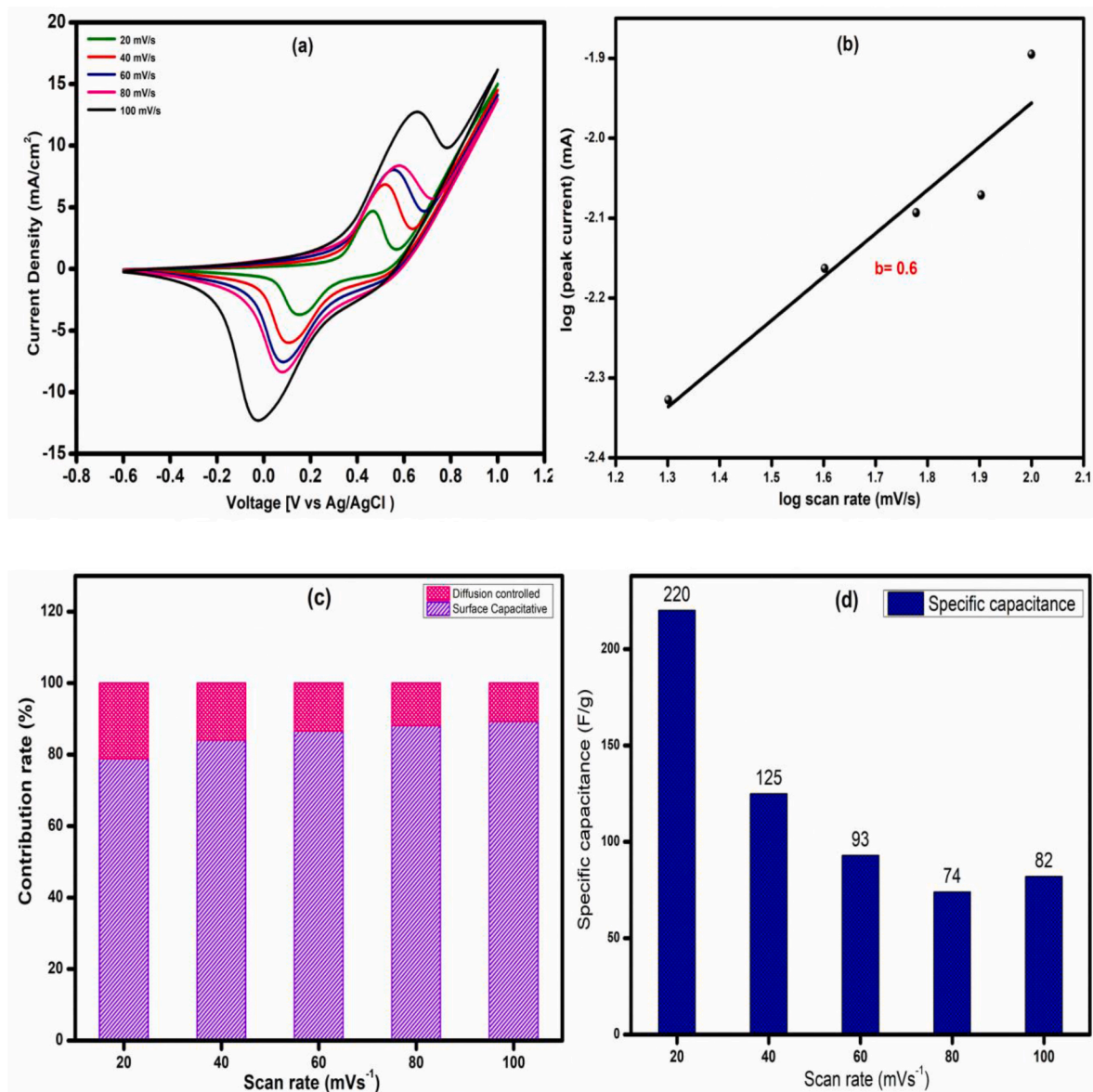


Fig. 6. (a) Cyclic voltammogram of SAS-NiO thin films with different scan rates, (b) log (i) against log (v) linear relationship, (c) Percentage of diffusion-controlled and surface capacitive behavior at different scan rates, and (d) Relationship between specific capacitance and scan rate.

3.3. XPS

XPS analyses were conducted to examine the stoichiometry and chemical characteristics of nickel oxide. As given in Fig. 5 (a), the survey scan spectra of NiO thin film confirms the purity and the presence of nickel and oxygen in the electrode. Fig. 5 (b and c) shows the high-resolution XPS spectra of the Ni (2p), and O (1s) core level obtained from NiO sample. The core-level spectrum exhibits a doublet associated with shake-up satellite peaks that corresponded to the Ni (2p) core values for NiO. The deconvoluted spectra include many peaks at 853.82, 854.59, 855.68, 857.98, 860.85, 864.33, and 866.62 eV, corresponding to Ni 2p_{3/2}. The peaks at 871.26, 872.77, 874.46, and 879.47 eV correspond to the Ni 2p_{1/2} core level. The detected peaks at 853.82, 854.59, 871.26, and 872.77 eV correspond to the Ni²⁺ state [45], indicating nickel–oxygen bonding and the face-centered cubic shape of the crystal structure [46]. The peaks detected at 855.68, 857.98, and 874.46 eV, corresponding to the Ni³⁺ state, which is ascribed to nickel vacancies [47]. Satellite peaks were detected at 860.85, 864.33, 866.62, and 879.47 eV [48]. The peaks associated with Ni³⁺ states exhibit a

large area, suggesting a higher Ni³⁺ concentration, which serves as an electrochromic active factor and enhances electrochromic performance [49]. Also, the presence of Ni³⁺ introduces extra redox-active sites, thereby enhancing electrochemical activity. Additionally, the existence of vacancies expands the potential window, facilitating larger colour tunability, as well as increases energy density by enhancing capacity and improving cyclic stability through enhanced electron transport properties [50]. The core level spectra of the oxygen 1s level Fig. 5 (c) exhibit two peaks at 529.6 eV, 531.5 eV, and 532.5 eV which are ascribed to the O–Ni (Ni²⁺) and O–Ni (Ni³⁺) states, O–H group or adsorbed oxygen, respectively [51].

3.4. Electrochemical measurements

The electrochromic characteristics of the NiO thin film deposited on the OMO films are investigated using a standard three-electrode electrochemical system. The system consisted of a NiO thin film deposited on the OMO as the working electrode, Pt as the counter electrode, Ag/AgCl as the reference electrode, and 1 M KOH as the electrolyte.

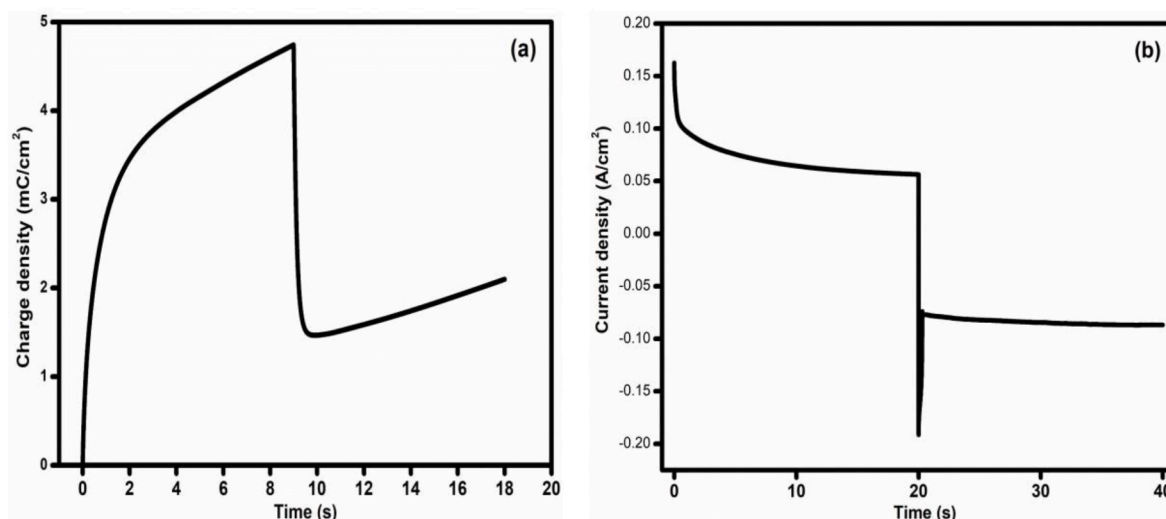
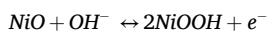


Fig. 7. (a) Chronocoulometry curve of SAS-NiO thin film, (b) Time response of the current density of SAS-NiO thin film.

Fig. 6 (a) depicts the cyclic voltammetry (CV) curve of NiO thin films with different scan rates ranging from 20 to 100 mV. The CV curves exhibit two strong redox peaks: the anodic peak and the cathodic peak, which occur at potential potentials of about 0.6 V and -0.02 V, respectively. When voltage is applied between -0.6 V and $+1$ V, an oxidation process occurs in which NiO combines with the OH^- ions in the KOH electrolyte and forms NiOOH. This reaction leads to the extraction of electrons from the film. Consequently, the film turns from bleached (transparent) state to colored (dark brown) state. Furthermore, when a voltage ranging from $+1$ V to -0.6 V is applied, a reduction process occurs. This process involves the injection of electrons into the film, resulting in the return of OH^- ions to the electrolyte. Also, the outcomes suggest that the electrons are easily transported through the OMO layer. Hence, NiOOH undergoes a transition into NiO, leading to the progressive reduction of the dark brown colour and reaching the desired transparency. The reaction mechanism involves the reversible process of insertion and extraction of OH^- ions [52].



Moreover, the redox peaks have a symmetrical nature, indicating a strong inherent reversibility of NiO thin films. Evidently, the current density increase as the scan rate increases, and all the cyclic voltammetry curves exhibit an identical shape, suggesting that the prepared film favour rapid redox processes. At higher scan rates, the CV curves exhibit more pronounced symmetry, suggesting improved high-rate responsiveness of the NiO film. Further, as the scan rate is increased there is a shift in the potential and current at the two peaks (redox) towards the positive and negative axes. The observed change may be ascribed to the large easily available surface area of the NiO thin films and the fast rate of ionic/electronic diffusion during the faradaic redox process [53]. The electrochemical kinetics of NiO films were examined to get insight into the rate of response between various states in the EC films. The distinction of faradaic and capacitive-controlled mechanisms may be determined by examining the CV data based on the equation [54, 55],

$$i = av^b$$

$$\log i_p = b \times \log v + \log a$$

where v represents the scan rate, a and b are variable parameters. Determining the value of b by analysing the slopes of the curve generated by graphing $\log(i)$ against $\log(v)$ (Fig. 6(b)) might elucidate the process of ion insertion/extraction. Here, the value of b is close to 0.5

represents diffusion -controlled process, b close to 1.0 represents a capacitive process, and b between 0.5 and 1 represents the combined mechanism. The obtained b value is 0.6, implying that the total charge capacity of NiO thin film is controlled by both capacitor-like and diffusion-dominated behavior.

To determine the precise contributions made by the diffusion-controlled ion intercalation (Faradaic) process as well as surface capacitive behaviour to the overall current, the currents at selected potentials may be divided into two components based on an equation as follows [54,55].

$$i(v) = k_1 v + k_2 v^{1/2}$$

Here $k_1 v$ and $k_2 v^{1/2}$ are the current value resulting from the capacitive and diffusion processes, respectively. By finding the values of k_1 and k_2 , we can determine the exact percentages of currents attributed to capacitive and diffusion-controlled process, respectively. Fig. 6(c) illustrate that, for the NiO thin film, the surface-capacitive contribution constituted 79 % at a scan rate of 20 mV/s. The value then gradually raised to 89 % when scan rates were progressively increased to 100 mV/s (Fig. 6 (c)). The surface capacitive process clearly predominates the current contributions for the NiO thin film.

The capacity of the deposited thin film may be evaluated by the specific capacitance of the active materials on the substrate, using the following formula.

$$C_s = \frac{\int IdV}{vmV}$$

Where C_s represents the specific capacitance of the active material (Fg^{-1}), I denotes the response current (A), v indicates the scan rate (mVs^{-1}), m refers to the mass of the active material, and V signifies the applied potential window (V). Fig. 6 (d) illustrates the relationship between estimated specific capacitance and scan rate. The deposited film shows high specific capacitance value of 220 Fg^{-1} for a scan rate of 20 mVs^{-1} and higher than the reported values [56–61]. The specific capacitance value decreases with increasing scan rate which is mostly associated with basic electrochemistry concerning the limitations of electron kinetics and ion transport inside the NiO electrode. Furthermore, electron transport and OH^- ion diffusion increase at low scan rates owing to the sufficient time afforded for their passage across the whole electrode material, hence enhancing the specific capacitance. Conversely, at increased scan rates, electron dynamics and ion intercalation in the NiO material are hindered due to its relatively deficient electrochemical or faradaic redox characteristics [62].

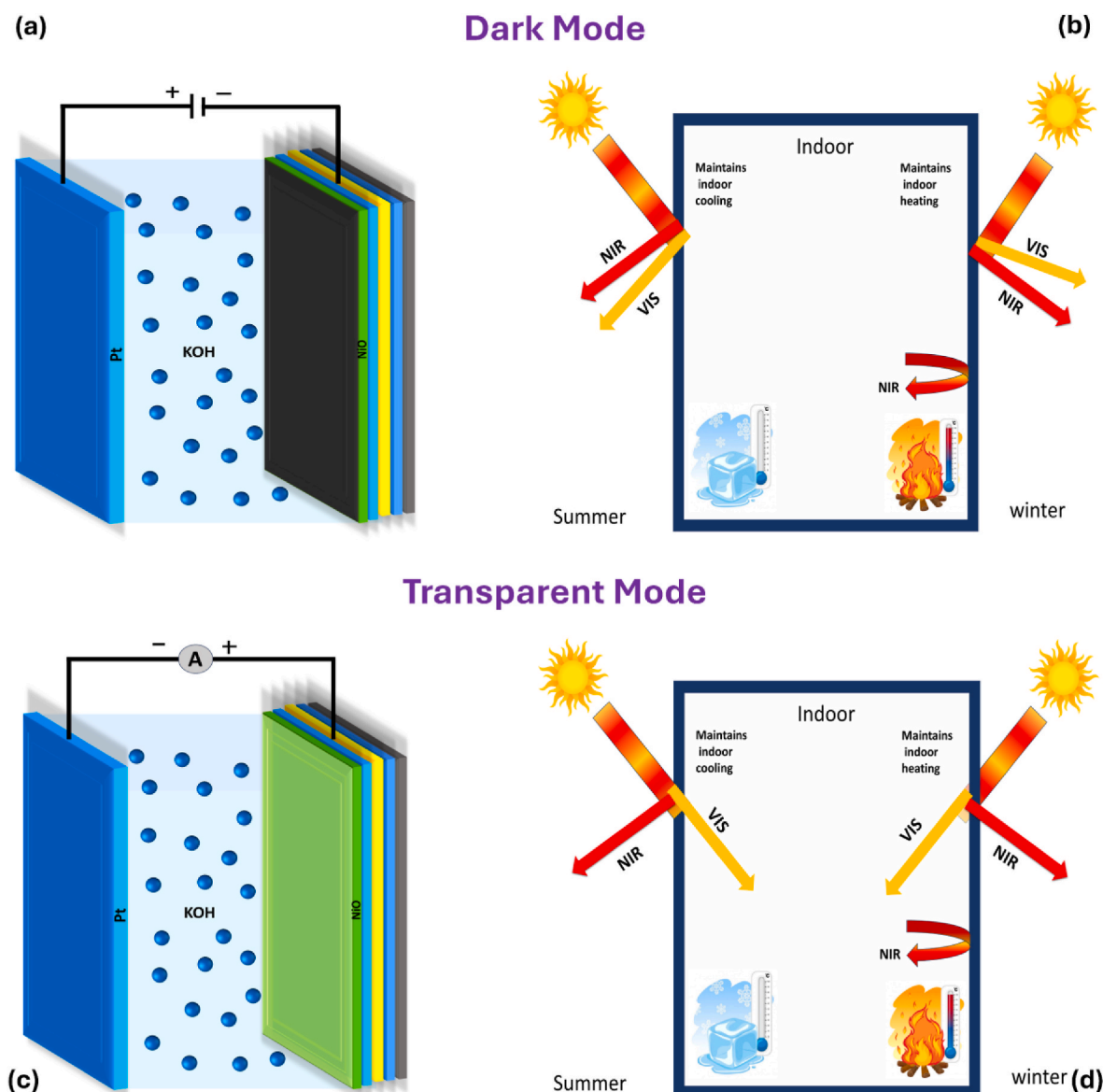


Fig. 8. a). The schematic illustration of Low-e electrochromic device with SAS-NiO electrode in the dark mode, (b) Low-e electrochromic window in dark mode during summer and winter, (c) Low-e electrochromic device with SAS-NiO electrode in the transparent mode, and (d) Low-e electrochromic window in transparent mode during summer and winter.

To further have a better understanding about the kinetics of ion intercalation in NiO film, we used the Randles-Sevcik formula [63] to compute the ion diffusion coefficient

$$i_p = 2.69 \times 10^5 \times A \times n^{3/2} \times D^{1/2} \times C \times v^{1/2}$$

here, i_p is the peak current (mA), A is the working electrode area (cm^2), n is the number of electrons transported, D is the ion diffusion coefficient ($\text{cm}^2 \cdot \text{s}^{-1}$), C is the ion concentration ($\text{mol} \cdot \text{cm}^{-3}$), and v is the CV scanning rate ($\text{mV} \cdot \text{s}^{-1}$). The anodic and cathodic peak currents exhibited linear correlations with the square root of the scan rate. This facilitated us to estimate the D value to be $2.19 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$.

CC provides quantitative data on the quantity of protons or ions intercalated (Q_i) or deintercalated (Q_{di}) with the application of a potential double step over a specified duration [64]. Fig. 7 (a) shows the CC curve for NiO thin films, taken at a potential window of $\pm 3 \text{ V}$, for 10 s with a charge intercalation of 4.76 mC/cm^2 . The quantity of deintercalated ions varies from that of intercalated ions due to the entrapment of some ions inside the sample, even after the cycle is reversed.

Hence, the reversibility is found to be 70 %. The response time for coloration (t_c) & bleaching (t_b) refers to the duration needed for the anodic/cathodic current to attain a steady-state level after the application of voltages, or the time necessary for the system to achieve 90 % of its total modulation. Fast switching responses indicate superior performance in electrochromic devices [64]. Fig. 7 (b) shows the reversible switching between the colored and bleached states taken at a potential window of $\pm 3 \text{ V}$, for 10 s. The switching durations are rapid, 3 s for coloration (t_c) and 1 s for bleaching (t_b). The electrochromic reaction time is influenced by the ion diffusion coefficient and the length of the dispersal channels [65]. The former primarily relies on the chemical and crystalline structure of the electrochromic materials, whilst the latter is dependent upon the microstructure of the electrochromic materials. Consequently, the electrochromic efficiency often relies on the surface morphology of the electrochromic materials [66,67]. The bleaching process takes place faster than the coloration process. In general, the reaction time for bleached forms is shorter and simpler than that of colored forms, owing to the lower conductivities of the bleached-state configurations compared to the colored-state configurations [68].

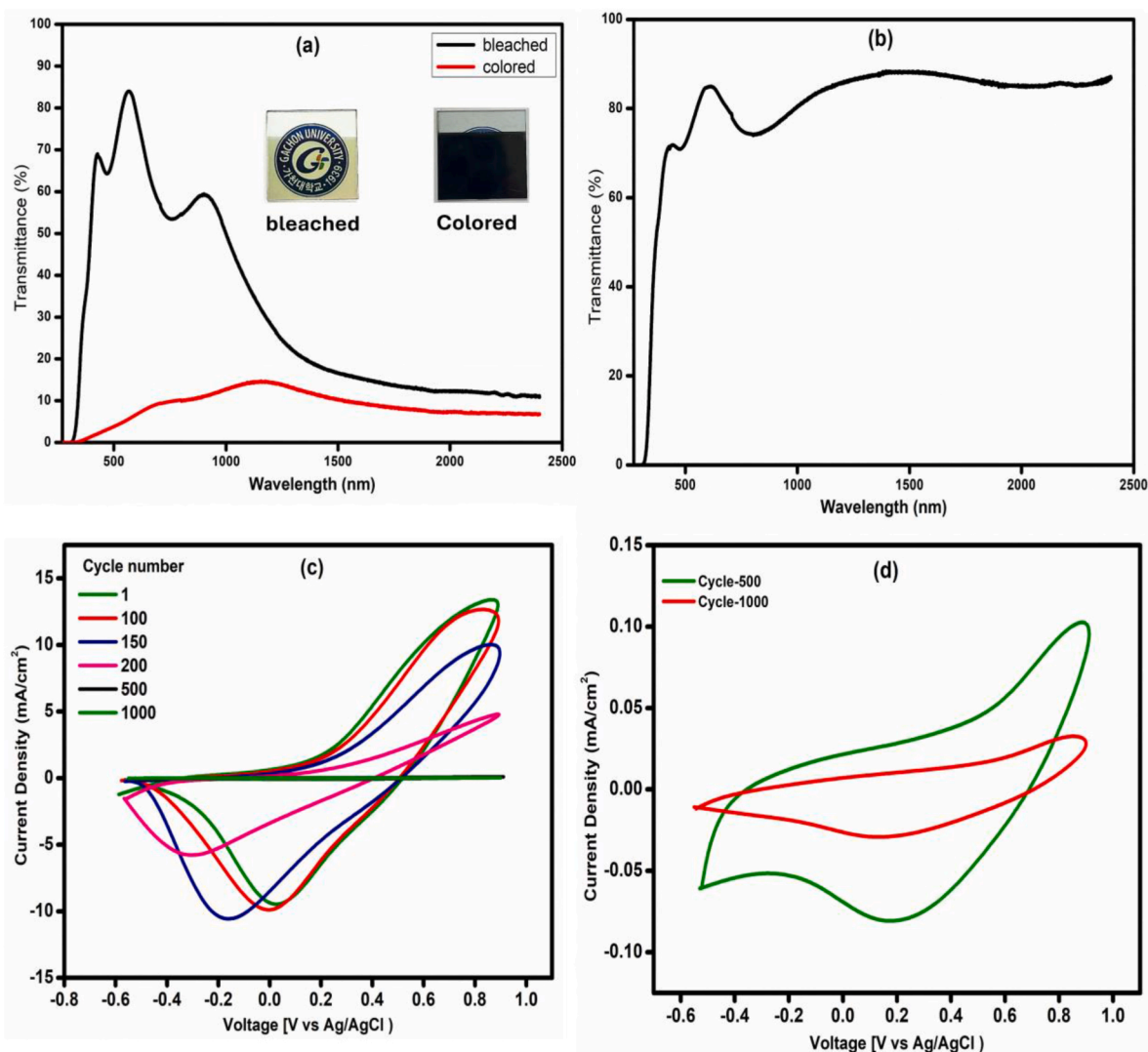


Fig. 9. (a). The transmittance spectra of Low-e SAS-NiO electrodes in the colored and bleached state with respective samples in the inset, (b) Transmittance spectra of NiO thin film deposited on glass substrate, (c) Cyclic voltammetry studies at different cycles, and (d) Cyclic voltammetry studies at 500 and 1000 cycles.

The optical transmittance measurements were carried out in two electrode setups by applying DC voltage of ± 3 V, with platinum as counter electrode, KOH as electrolyte, and the prepared SAS-NiO thin film as the working electrode. The schematic illustration of Low-e electrochromic device and their respective window model in the dark and transparent mode is shown in Fig. 8(a–d). Generally, during summer, around 50 percent of the solar heat that strikes a window will be transmitted inside. Utilizing a Low-e electrochromic glass as a window coating would reduce the interior heat transfer to 30 %, hence aiding in the preservation of cooler indoor environments throughout summer, as well as having high transparency to visible light. As well as, during winter, the Low-e electrochromic glass would save interior warmth by blocking internal heat escaping through the window to the outside environment and can function as a defroster to preserve window transparency when supplied with a DC voltage. Fig. 9 (a) shows the transmittance spectra of Low-e NiO film in the colored (inset of Fig. 9 (a)) and bleached states (inset of Fig. 9 (a)). For comparison the transmittance spectra for the NiO thin film deposited on glass substrate at similar condition were given in Fig. 9 (b). When the SAS-NiO film is applied with a potential of -3 V, it shows dark mode (Fig. 8 (a)) and becomes almost black colored where it blocks of 95 % of visible light and almost 88 % in the NIR region (Fig. 8 (b)). Therefore, the dark mode may greatly lower the energy consumption of the building for air

conditioning and lights. When the potential is reversed to $+3$ V, the NiO film becomes transparent (Fig. 8 (c)) exceeding 80 % over a broad range of frequencies across the visible spectrum, and blocks most of NIR (56 % (@ 950 nm, and 93 % (@ 2400 nm) (Fig. 8 (d)). Thus, conserving indoor warmth during winter by inhibiting the escape of internal heat to the external environment and conserving energy. The SAS-NiO electrode exhibited optical modulation of 78 % at 550 nm (VIS), 44 % at 950 nm (NIR), and 3 % at 2400 nm (NIR). Electrochromic cycle stability is a vital property that determines the durability of electrochromic devices. To investigate the cyclic stability of NiO film coated on SAS substrate in the KOH electrolyte and to study the peak current values during the coloration and bleaching process cyclic voltammetry studies were carried out for 1000 cycles. As given in Fig. 9 (c), the current values started to reduce slightly after 150 cycles and reduces more after 500 cycles. The CV curves at 500 and 1000 cycles are shown in Fig. 9 (d). The decrease in the peak current values with an increasing number of cycles is due to the degradation of NiO [69]. The low-density $\text{Ni}(\text{OH})_2$ incorporates water molecules during the oxidation process with OH, leading to degradation owing to hydration. Also, the degradation of NiO in aqueous electrolytes is associated with the partial dissolution of low-stability NiOOH during the oxidation process, along with subsequent self-discharge phenomena [70].

4. Conclusions

In summary, we developed a room temperature Low-e electrochromic electrode for the fabrication of ECDs. A multilayered structure of OMO, comprising SIZO/Ag/SIZO layers, is fabricated at room temperature using the sputtering process as a TCO material. This material has an amorphous structure with high visible transmittance of 97 % at 550 nm, and very low transmittance in NIR region (54 % at 950 nm, and 5 % at 2400 nm). Additionally, its sheet resistance is 4 $\Omega/\text{sq.}$, far lower than that of commercially available ITO. NiO thin films were deposited at room temperature on SAS multilayers. XRD confirms the amorphous nature. FESEM depicts triangular pyramid like morphology with some voids present in it. XPS spectra revealed the presence of large concentration of Ni^{3+} states along with Ni^{2+} states, which creates more redox-active sites and increases electrochromic performance. The SAS-NiO electrode exhibited exceptional electrochromic performance, with optical modulation of 78 % at 550 nm (VIS), 44 % at 950 nm (NIR), and 3 % at 2400 nm (NIR), in addition to notable cycle stability. The developed SAS-NiO film can keep the home cool in the summer and warm in the winter, allowing for comfortable interior weather and clear outdoor vision without haze. The capacitive electrochromic electrode, with a specific capacitance of 220 Fg^{-1} , offers enhanced electrochemical kinetics, significant charge capacity, and is appropriate for use as a secondary energy storage device. Hence, we hope that the designed electrochromic low-emissivity films for building wall thermal envelopes provide a solution for year-round energy efficiency in heating and cooling.

CRediT authorship contribution statement

K.S. Usha: Writing – review & editing, Writing – original draft, Data curation, Conceptualization. **Sang Yeol Lee:** Writing – review & editing, Validation, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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