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Copper Oxide–Based MOS Capacitors for High-Frequency Applications

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Abstract

Cu₂O is an important material for optoelectronics and communication technologies because of its suitable bandgap, strong light absorption and potential for low-cost device fabrication. Improving its crystallinity is essential to reduce defects, enhance charge transport and achieve better optical and electronic performance, leading to more efficient and reliable devices. Herein, copper oxide thin films deposited onto glass and n-Si silicon substrates and partially recoated with molybdenum nanosheets were employed as high frequency metal-oxide-semiconductor (MOS) capacitors. As a result, Mo-coating improved the crystallinity and lowered the defect density in CuO. Optically, Mo nanosheets widened the energy band gap without significant variation in absorption spectra of the glass/CuO films. The MOS capacitors also displayed accumulation-depletion-inversion switching characteristics in the frequency domain of 1.0-100 MHz allowing for ultrafast charging/discharging cycle within 10 ns. In addition, Mo-coating lowered the built in potential and tuned the accumulation-inversion switching mode. Moreover, treating the MOS devices as high frequency bands filters in the range of 1.0 M- 6.0 GHz showed the ability of the devices to transmit/reject signals in three high frequency bands centered at 1.02 GHz, 1.47 GHz and 3.30 GHz. The ideal reflection coefficient values approaching zero, return loss characteristics exceeding -43 dB and voltage standing wave ratios (VSWR) equal 1.0 were obtained at notch frequency of 3.30 GHz; the bandwidth at this frequency reaches 2.0 GHz. The features of the CuO-based MOS capacitors presented by ultrafast mode switching, high frequency filtering, wideband-frequency control, large values of return loss and ideality of VSWR nominated them for 6G technology applications.

Keywords: n-Si/p-CuO, Mo nanosheets, MOS, Band filter, 6G, High frequency.

1. Introduction

Copper oxide thin films attracted the interest of researchers and industry sector due to their smart applications and CeO₂-doped CuO performed as supercapacitors exhibiting areal capacitance of 0.14 F/cm² [1]. These capacitors displayed superior electrochemical stability showing retention of 100% and displayed Coulombic efficiency of 99.4% [1]. More specifically, CuO based solar cells showed open circuit voltage of 0.59 V, short circuit current density of 16 mA/cm² and filling factor of 60% indicating a promising characteristic for future solar cell applications [2]. In addition, hydrothermally grown copper oxide thin films illustrated memristive phenomenon presented by pinched hysteresis loop applicable as resistive random access memories [3]. Moreover, copper oxide thin films deposited onto Si substrates by chemical spray pyrolysis technique exhibited photodetectors characteristics showing current responsivity of 2.85 A/W, external efficiency of 279 % and specific detectivity of 2.7×10^{12} Jones when exposed to visible light illumination [4]. As an important feature, CuO was employed to enhance the reception of terrestrial broadcasting and integrated serviced digital broadcasting in the 470-770 MHz frequency band [5]. CuO modified antennas showed average gain of -1 dBi at notch frequency of 600 MHz [5]. The bandwidth of this device reached 300 MHz.

In spite of its important technological applications, copper oxide demonstrated short diffusion lengths between 100 nm and 200 nm [6]. Such a property prohibits charge separation process leading to photodegradation effects [6]. Copper oxide suffers from rapid capacity fading at high levels of charging/discharging rates negatively affecting the charge storability [7]. Different physical and chemical methods were employed to enhance the electrical conductivity and chemical stability of CuO thin films [7]. One of these methods employed the molybdenum as a doping agent to enhance the electrical properties of CuO [8]. Mo doping enhanced the electrical conductivity and specific capacitance as well. Mo-doped CuO nanosheets exhibited specific capacitance of 1392 F/G at 2A/g, revealed energy density of 36 Wh/kg at 810 W/kg and displayed cycle life of 81% after retention of 5000 cycles.

The important applications of CuO in wide range of technology and the enhancements achieved via Mo-corporation into the structure of CuO represented a motivation for the production of a multichannel device that can perform as Metal-Oxide-Semiconductor (MOS) capacitors and as high frequency band filters. Particularly, the work here was concerned with the production of multichannel device made of CuO films deposited onto n-Si substrates. One of the channels was coated with Mo nanosheets of thickness of 100 nm to enhance the electrical performance as suggested in literature [8]. The produced devices were studied by means of X-ray diffraction, impedance spectroscopy and network analysis spectroscopy. The effect of Mo nanosheets on the accumulation, depletion and inversion properties of the capacitance -voltage characteristics in the frequency domain of 1.0-100 MHz was explored to determine the shortest possible charging/ discharging time scale so that the constructed MOS device could be utilized in high frequency applications. In addition, the band filtering characteristics of the device channels were tested in the frequency domain of 1.0 M-6.0 GHz to determine the notch frequency. The pass/reject bands and their return loss value and voltage standing wave ratios were investigated so that their suitability for 6G applications could be determined. As an important information needed to proof the role of the energy band gap widening on the MOS device performance, the optical transmission, reflection and absorption of the CuO and CuO/Mo structures were studied and discussed.

2. Experimental details

Copper oxide thin films ($1.0\ \mu\text{m}$) were deposited onto ultrasonically and chemically cleaned glass and n-Si substrates ($300\ \mu\text{m}$ thick wafers) using NORM-600 VCM thermal evaporator under a vacuum pressure of 10-5 mbar. The thickness of the CuO films was in situ monitored using Inficon-STM-2 thickness monitor installed in the thermal evaporator. The substrate temperature was 300 K and no annealing process were carried out. The source material was CuO powders of 99.99% purity ordered from Alfa Aesar firm. Some of the produced glass/CuO and n-Si/CuO films were masked (square masks) and installed in an ion coater to deposit a molybdenum layer of thickness of 100 nm. The Mo target was of very high purity (99.99%, Alfa Aesar). The thickness of the nanosheets was determined by a SOLID INC. II profilometer. Some of the n-Si/CuO and n-Si/CuO/Mo films were point contacted with Ag (99.999%) point contacts to fabricate capacitors of contact area of $0.0314\ \text{cm}^2$. The rear side (n-Si) of the device arrays was coated with Ag. The schematic of the device was shown in inset-1 of Fig. 1. The structural properties of the produced films were explored using Miniflex 600 XRD unit, and the capacitance-voltage characteristics were recorded in the frequency range of 1.0-100 MHz using an Agilent 4291B-1.0M-1.80 GHz impedance analyzer. The magnitude of the reflection coefficient spectra in the frequency domain of 1.0M-6.0 GHz was measured using Rohde & Schwarz network analyzer 9 kHz-6 GHz. The optical spectra were measured using Thermo-Scientific Evolution 350 spectrophotometer equipped with VEE-MAX-II PIKE variable angle reflectometer. The p-conductivity type of CuO was measured by the hot probe technique.

3. Results and Discussion

The current study targeted the design and fabrication of high frequency ($\sim 100\ \text{MHz}$) depletable metal-oxide-semiconductor (MOS) capacitors made of copper oxide thin films deposited onto n-Si wafers. The design considered the presence of two channels. One was as it was grown composed of n-Si/Cu₂O (abbreviated as SCM-00) and the other was n-Si/CuO top coated with 100 nm thick Mo nanosheets (SCM-100). The schematic for the two channels is shown as inset-1 in Fig. 1. Fig. 1 also shows the X-ray diffraction patterns for the n-Si substrates, n-Si coated with CuO and n-Si/CuO coated with Mo nanosheets. Better details about the observed patterns are presented as inset-2 in Fig. 1. While SCM-00 films displayed very broadened peak centered at $2\theta = 31.78^\circ$ and assignable to the monoclinic phase of CuO ($a = 4.83\ \text{\AA}$, $b = 3.76\ \text{\AA}$, $c = 5.74\ \text{\AA}$, $\beta = 99.18^\circ$ and space group of C_2 (group no: 5)), films coated with 100 nm thick Mo nanosheets displayed much sharper and narrower peak centered at $2\theta = 32.85^\circ$. Both of the peaks were assigned to the same crystal structure along the (111) direction for the peak of SCM-00 and along (110) for SCM-100. The lattice parameters used to explore the observed XRD patterns were partially related to the international center for diffraction data PDF card no: 05-0661). They were also consistent with literature data [9].

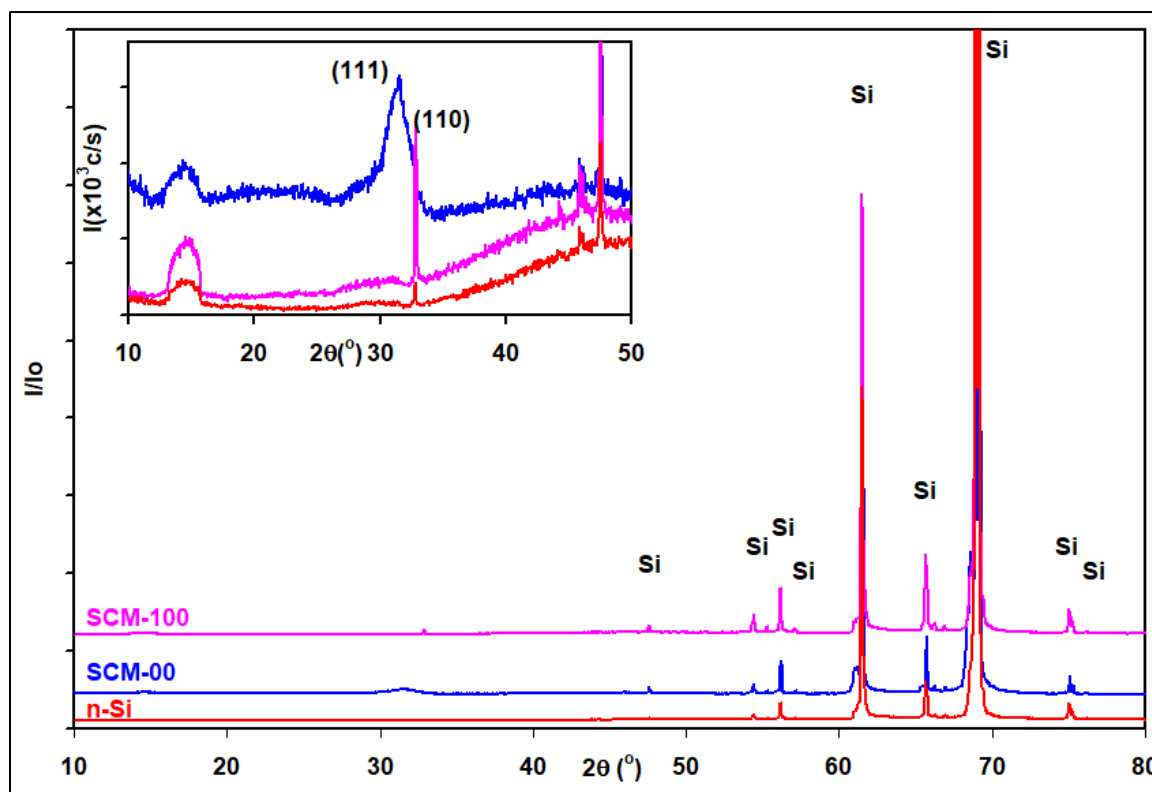


Figure 1. The X-ray diffraction patterns for Si/CuO and Si/CuO/Mo films. Inset-1 showing the schematic for the device and inset-2 showing an enlargement of the XRD patterns in the low diffraction angles ranges (25-40 o).

Most of the peaks relating to the monoclinic structure were absent from the XRD profiles due to the high degree of disorder in the CuO layer. Attempts to explore the role of Mo nanosheets on the structural properties of CuO included calculation of the crystallite size ($D = 0.94\lambda/(\beta \cos(\theta))$), the microstrain ($\epsilon = \beta/(4 \tan(\theta))$) and the dislocation density ($\delta_c = 15\epsilon/(aD)$) alongside one of the crystal axis [10]. Mo nanosheets increased D and decreased ϵ and δ_c values of CuO from 5.0 nm, 2.85×10^{-2} and 1.59×10^{13} lines/cm² to 43 nm, 2.96×10^{-3} and 1.78×10^{11} lines/cm², respectively. The remarkable enhancement in the crystallite size, decreased in microstrain and in defect density by two orders of magnitude, indicated that Mo nanosheets played a vital role in the structural properties of CuO films. The most probable reason for these enhancements was the ionic substitution of Mo+6 (ionic radius is 62 pm [11]) or Mo+4 (68 pm [11]) in sites of Cu+2 (73 pm [12]). Diffusion of Mo ions in sites of CuO, whose bond length was 192.8 pm [13], established bonding between Mo and O forming shorter bonds of Mo-O (bond length is 182.5 pm [14]). Increased number of bonds decreased the defect density because the defects mostly originate from Cu cation interstitials or oxygen atoms occupying four coordinated sites in CuO [15]. The crystallinity enhancements, achieved via Mo nanosheets, resulted in improved carrier confinement within the active region of electronic device [16]. The confinement enhanced the efficiency of charge carrier transport leading to higher carrier mobility [16].

Fig. 2 (a), (b) and (c) show the capacitance (C)- voltage (V) characteristic curves for the device under study (DUT) being recorded at various frequencies in the range of 1.0-100 MHz. It is evident from the figures that the DUT can be depleted showing a frequency and voltage dependent depletion regions marked by pinky shadows in Fig. 2(a)-(c). The slope of the $C - V$ curve in that region was sharper for SCM-100 channels than that of SCM-00. Notably, the higher the imposed ac signals frequency are the less the difference between the

accumulation and inversion regions and the narrower the depletion range are. For a standard MOS device, the surface potential $\phi_s \approx 0.0$ eV in the accumulation region, where the applied voltage, was less than the flat band voltage (V_{fb}) [17]. As the applied voltage exceeded V_{fb} (shown in Fig. 2 (a)), the device was in the depletion region and the surface potential increased with increasing applied voltage reaching a value of $\phi_s = 2\phi_b$ at the inversion region ($V = V_t$ in Fig. 2 (a)) [17]. With increasing ac signal frequency, the accumulation and inversion regions widened, whilst the depletion region narrowed. Numerically, for SCM-100 channel, where the $C - V$ variation, was more pronounced, V_{fb} exhibited values of -0.72 V, -0.45 V and -0.38 V whilst V_t exhibited values of $+0.49$ V, $+0.28$ V and $+0.13$ V, at signal frequency values of 1.0 MHz, 10 MHz and 100 MHz, respectively. Physically, negative applied voltage leads to flow of more holes that accumulate on the surface of the channel till the applied voltage become slightly positive repelling holes hence forming a depletion region with few free charge carriers. More positive voltage makes the flow of the minority carriers (electrons) more dominant at the surface leading to the inversion mode. Remarkably, this behavior is important for the onset of the digital MOS field effect transistors (MOSFET). MOSFET is “ON” and current flows between drain and source when the device reaches the inversion mode. The high frequency operation mode reaching 100 MHz indicates high switching speed. Practically, this means the MOS device at 100 MHz which corresponds to 10 ns period of time must charge/discharge every 5 ns per half cycle. Such property makes the device suitable as $5G/6G$ radio frequency amplifiers and in digital circuits the support high speed digital logic “CPU” unit. SCM-100 channels with these features can also be nominated for fast voltage-controlled tuning in antennas and band filters [18].

In order to understand the physics laying beyond the performance of the device, an abrupt depletion approximation analysis was held assuming the validity of the equation [17],

$$C^{-2} = \frac{2(V - V_{bi} - \frac{kT}{q})}{qA\epsilon_r N} \quad (1)$$

Here $A = 0.0314$ cm² is the contact area, V_{bi} is the flat band potential and ϵ_r is the dielectric constant. $\epsilon_r = 29.5$ and 17.1 for SCM-00 and SCM-100, respectively, was measured by an impedance analyzer at 1.0 GHz (high frequency limit). N is the reduced charge carrier concentration ($N = \frac{N_A N_d}{N_A + N_d}$; N_A , N_d are the acceptor and donor concentrations, respectively). Representative linear fitting of the $C^{-2} - V$ variation at 1.0 MHz is illustrated in Fig. 2 (d). The linear plots allowed calculation of the built in potential and the reduced charge carrier density as function of signal frequency. The frequency dependent V_{bi} and N is illustrated in Fig. 2 (e) and (f), respectively. For SCM-00 channel, the figures indicated a slow decrease in the built in potential with increasing signal frequency in the range of 1.0 - 50.0 MHz. For the same channel, similar behavior of $N - f$ variation was also observed in the frequency range of 2.0 - 100 MHz. The built in potential increased from 1.06 eV to 1.36 eV as the frequency increased from 50 MHz to 100 MHz. For this channel, the inability of the free charge carriers to follow the ac signals lay beyond the decrease in N and increase in V_{bi} at high frequencies (50 - 100 MHz). On the other hand, SCM-100 channel (Fig. 2 (e) and (f)) show an increasing V_{bi} and N with increasing signal frequency. Further, the value of the built in potential in SCM-100 channel was much less than at the SCM-00 channel. It was also noted that the value of N decreased upon coating the surface of CuO with Mo nanosheets.

The effect of Mo nanosheets as a doping agent presented by diffusion of Mo ions into sites of Cu+2 led to the decrease in the built in potential at a particular frequency. The relation of the work function of CuO (ϕ_{CuO}) to the reduced carrier concentration N could be read from the equation [17],

$$\phi_{CuO} = \chi_{CuO} + \frac{E_g}{2} + \frac{kT}{q} \ln\left(\frac{N}{n_i}\right) \approx 5.40 \text{ eV} \quad [19] \quad (2)$$

$$V_{bi} = |\phi_{Mo}(4.6 \text{ eV}) - \phi_{CuO}(5.40 \text{ eV})| = 0.80 \text{ eV} \quad [20] \quad (3)$$

Here, $\chi_{CuO} = 4.07 \text{ eV}$ [19], $E_g = 2.05 \text{ eV}$ (measured in the laboratory) and n_i is the intrinsic carrier concentration in CuO. Eqn. (2) and Eqn. (3) indicated that $V_{bi} \propto -\ln(N)$ which means the doping should increase N . However, this is not the case observed in Fig.2 (f). Even after Mo coating, N of Si/CuO is still larger than Si/CuO/Mo structure. Considering Mo+6 ions replacing Cu+2 in p-type CuO, then excess four electrons are donated into CuO [21]. As the defect density decreased (XRD analyses) owing to the filling of copper vacancies and completed bonding of Mo with oxygen interstitials, adding electrons to the p-type CuO compensates holes, converts the material to n-type and introduce localized states that in turn narrows impurity bands [22]. Narrowing of the impurity bands results in an increase in the energy band gap width [22].

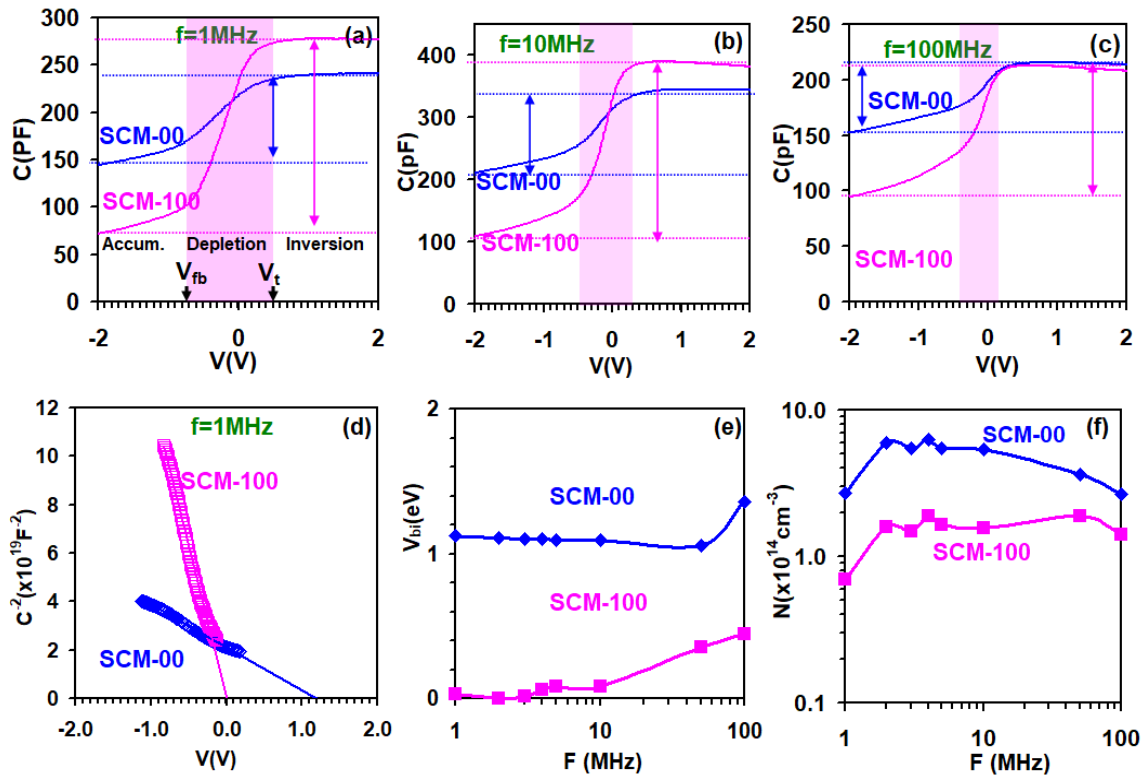


Figure 2. the capacitance-voltage characteristics for CuO-based devices measured at (a) 1.0 MHz, (b) 10.0 MHz and (c) 100 MHz. (d) are the abrupt depletion analyses presented by $C^{-2} - V$ variation for one of the $C - V$ curves and the frequency dependence of (e) the built in and (f) the reduced carrier concentration of CuO based MOS devices.

The band gap widening was confirmed from the absorption coefficient spectra and the related analyses, which are presented in Fig. 3 (a) and (b). The absorption coefficient (α) spectra, which were determined from the measured transmittance (T) and reflectance (R) spectra for films coated onto glass substrates (abbreviated as GCM-00, GCM-100; G is glass) to reserve light transmission were calculated from the relation [10],

$$T = (1 - R_{glass})(1 - R_{CuO})(1 - R_{CuO/Mo}) \exp(-\alpha d) \quad (4)$$

Here d is the total film thickness. T and R spectra displayed in Fig. 3 (a) indicated slight effect of Mo nanosheets on the transmittance and reflectance spectra of CuO. T spectra were slightly redshifted, whilst reflectance spectra were blue-shifted showing larger R values for GCM-100 films. The absorption coefficient spectra, which are illustrated in Fig. 3 (b), showed large decay of α values with decreasing incident photon energy exhibiting a local absorption peak centered at photon energy of 1.79 eV and 1.71 eV for GCM-00 and GCM-100, channels, respectively. The local peak and the blue shift in α spectra were better visible from the inset of Fig. 3 (b). This peak was different from the peaks appearing in T spectra at 2.18 eV and in R spectra at 1.61 eV and 1.68 eV for GCM-00 and GCM-100, respectively. The maximum transmittance value occurring at $E = 2.18$ eV corresponds to the limit of “yellow” excitonic series in Cu₂O revealing optical transitions originating from $\Gamma_{7v}^+ - \Gamma_{6c}^+$ of the first Brillouin zone of Cu₂O [23]. The energy values near 1.60 eV were attributed to the oxygen vacancies situated at the surface of the films [24]. The energy values revealing local absorption maxima at 1.79 eV correspond to transitions in CuO rather than Cu₂O [25]. On the other hand, the Tauc's equation $((\alpha E)^2 \propto (E - E_g))$ [10]) fittings illustrated in Fig. 3 (c) indicated that the E –axes crossings revealed an energy band gap of value of 2.05 eV that blue-shifts to 2.20 eV upon coating of CuO with 100 nm thick Mo nanosheets. Hence, by experiment, the energy band gap widened because of Mo diffusion resulting in an increase in the concentration of electrons and forcing compensation of holes.

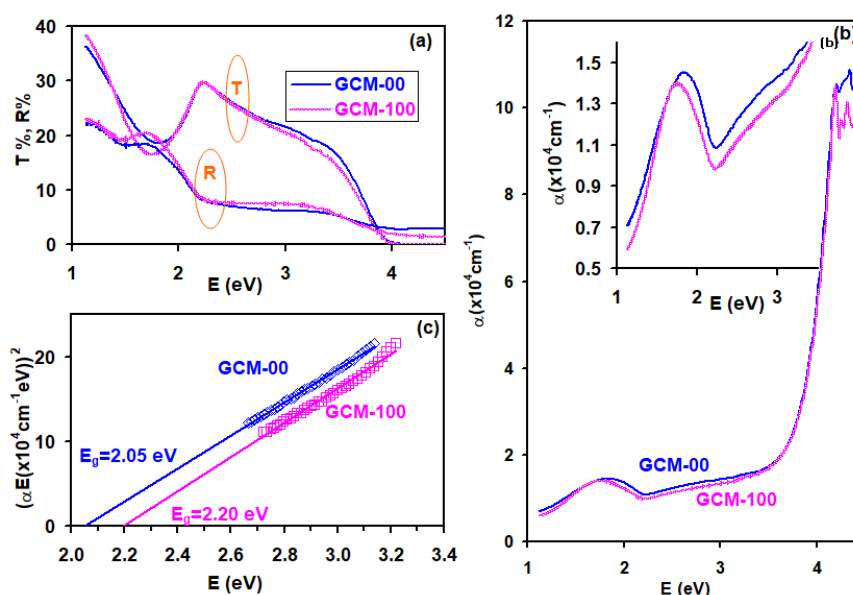


Figure 3. (a) the transmittance and reflectance spectra, (b) the absorption coefficient spectra and (d) the Tauc's equation fittings for glass/CuO films before and after coating with Mo nanosheets. Inset of (b) showing the low absorption range.

In addition to the ultrafast charging/discharging facility of the SCM-xx devices suiting 6G technology applications, the electrodes of the device channel were imposed between the terminals of a Rohde & Schwarz R&S ZVL network analyzer workable in the range of 9 kHz to 6 GHz. The measured absolute value of microwave reflection coefficient “ S_{11} ” spectra are illustrated in Fig. 4 (a). It is evident from the figure that a good correlation between the S_{11} parameters of both channels existed. Three main minimal peaks were detected in the spectra of SCM-00 channels, namely S_{11} of SCM-00 exhibited values of 0.146, 0.17 and 0.02 at 1.02 GHz, 1.47 GHz and 3.27 GHz, respectively. Coating the channel with 100 nm Mo nanosheets increased the respected values to 0.121, 0.189 and 0.0074. The absolute minima detected at 2.27 GHz in SCM-00 were

observed at 3.30 GHz in the S_{11} spectra of SCM-100 channels. The return loss ($L_r = 20 \log(S_{11})$ [26]) spectra for the DUT are displayed in Fig. 4 (b). The first local minima observed at 1.02 GHz and 1.47 GHz exhibited L_r values of 16.6 dB and -15.6 dB, respectively, whilst the absolute minima centered at 3.27 eV exhibited L_r value of -32.4 dB. Coating the CuO layer with Mo nanosheets increased the return loss values at 1.02 GHz and at 3.30 GHz to -18.3 dB and -42.59 dB, respectively. In addition, the voltage standing wave ratios ($VSWR = \frac{1+S_{11}}{1-S_{11}}$) spectra presented in Fig. 4 (c) revealed values of 1.28, 1.46 and 1.01 at 1.20 GHz, 1.47 GHz and 3.30 GHz, respectively.

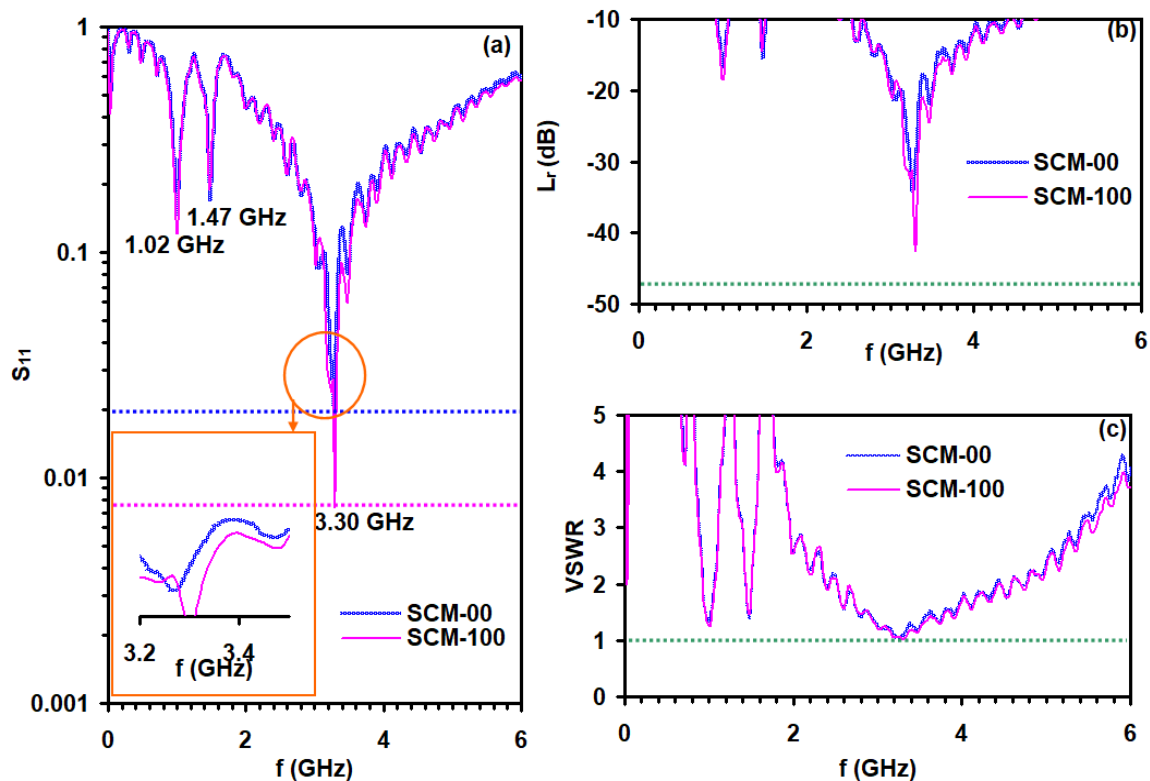


Figure 4. (a) the magnitude of the reflection coefficient spectra, (b) the return loss spectra and (c) the voltage standing wave ratios for SCM-xx devices being studied in the microwave frequency range of 1.0 M-6.0 GHz. Inset of (a) showing the center frequency shift of the absolute minima.

S_{11} parameter represents the value of the power reflected back at the notch frequency (stop band frequency). The return loss parameter measures the power that is not absorbed by the SCM-xx device and therefore is returned to the network analyzer. Acceptable values of L_r exceed -10 dB whilst excellent values are assumed to be less than -20 dB [26], [27]. $VSWR$ values represent the peak voltage that can be found on a line under non-ideal match conditions. It measures the “goodness” of the match [27]. Excellent impedance match and full signal transition occur at S_{11} , L_r and $VSWR$ values of 0.0, -20 dB and 1.0, respectively. The values were recorded at the notch frequencies 3.27 GHz and 3.30 GHz, for SCM-00 and SCM-100, respectively, which are also shown in Table-I, indicated that the proposed SCM-xx channels are ideal for 5G/6G communication technologies. Based on -10 dB thresholds, the bandwidths (BW) of the peak detected at 3.30 GHz was 2.0 GHz. It indicated that the DUT covers a large frequency span. This is a property of wideband notch filter employable in multiband 5G/6G radios, Radar and jamming resistance [28] and EMI (electromagnetic interference)/RFI (radio frequency interference) suppression [29].

It is evident from table-I that SCM-00 and SCM-100 device channels are one of the most impressive channels employable in 5G/6G technology applications. The filter model named RF-Lambda RNFL3351K and the filter model NEDITEK- NDMF02001 is commercial filters sold in market and designed as cavities for 5G and near 6GHz filtering and for 5G interference suppression of C band, respectively, exhibited filter parameters highly combatable with the currently reported device channels indicating the novel properties of the currently proposed devices. The currently proposed devices show the widest bandwidth ever seen in this technology sector.

Table 1. show some of the band filters employed in high frequency technology.

Filter Model	Center Freq (GHz)	S_{11}	Return Loss [dB]	VSWR	Bandwidth (GHz)	Notes	Ref .
RF-Lambda RNFL3351 K	3.35	0.0024	54.2	1.007	0.234	Cavity notch for 5G NR sub-6 GHz filtering	[30]
NEDITEK NDMF02001	4.6	0.006	45.0	1.017	0.100	Band-stop for 5G interference suppression (C-band)	[31]
Square open loop resonator	2.4	0.316	10.0	1.09	0.56	WLAN/Bluetooth/Zigbee/RFID/ISM applications	[32]
MIMO array antenna	3.56	0.33	9.54	2.0	0.68	5G smartphones and IoT	[33]
SCM-00	3.27	0.02	34.21	1.06	2.0	6G technology	*
SCM-100	3.30	0.007	42.59	1.02	2.0	6G technology	*

*Current work

4. Conclusions

In the current work, the researchers showed the possible fabrication of novel metal-oxide –semiconductor devices employable as ultrafast rechargeable capacitors and as band stop filters. The physical design of the devices included coating of copper oxide layer with molybdenum nanosheets to enhance their conductivity. The structural investigations showed the induced crystallinity of copper oxide via Mo nanosheets coating resulting in reduced defect density by two orders of magnitude. The optical analysis showed that Mo coating widens the energy band gap without affecting the absorption spectra of the oxide films. It was observed that the device under study could be depleted at high frequency reaching 100 MHz. In addition, as bandstop filters the uncoated and Mo coated channels exhibited smart bandstop characteristics presented by ideal reflection coefficient, ideal return loss parameter and ideal voltage standing wave ratios. The respective values reaching 0.007, 42.59 dB and 1.02 at a center frequency of 3.30 GHz with ultra-wide bandwidth made the Si/CuO/Mo devices compatible with those released in 2025 for 5G/6G applications. The currently reported device exhibited much wider bandwidth than those used in electronic markets.

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6. Declarations

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Conflict of interest/Competing interests

The authors declare that they have no conflict of interest

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Author(s) Contribution

Prof. A.F. Qasrawi, prepared literature review, guided the work, edited the article explored all parts of qualitative research. He also analyzed the optics and the XRD parts. Prof H. Khanfar designed a MATLAB based software packages to evaluate the transport parameters and to register the impedance spectra from the analyzer memory.

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