Study of dielectric properties in stacked high-k dielectric MOSCAP (Metal Oxide Semiconductor Capacitor) devices.

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Abstract

With the current technology having reached the physical limits of size downscaling, efforts need to be made toward the development of new dielectric materials to ensure a low leakage current density that can prevent the tunneling of charges through the dielectric layer. The tunneling of the charges through the dielectric layer is a major factor for degradation in performance as well as higher power consumption. Therefore, recent research has been focused on using materials with high dielectric constants and low leakage characteristics. In this work, we present a detailed study on ZrO_2 and Ta_2O_5 as potential high-k dielectric replacements. MOSCAP devices fabricated using high-k materials have been investigated to study their dielectric properties. Furthermore, a stacked bilayer structure of ZrO_2 and Ta_2O_5 was also investigated to alleviate some of the issues that come along with using high-*k* dielectrics instead of the more conventional SiO_2 dielectric thin film. ZrO_2 and Ta_2O_5 dielectric thin films were successfully fabricated on N^{++} doped silicon substrates using the electron beam evaporation technique displaying dielectric constants of 20 and 34, respectively. The fabricated MOSCAP devices exhibited leakage current densities as low as 10^{-8} A/cm^2.

Introduction

SiO_2 has been the dielectric of choice for modern electronics in devices such as metal oxide semiconductor field effect transistors (MOSFETs), metal oxide semiconductor capacitors (MOSCAPs), and thin film transistors (TFTs). The ability to be conformally coated over silicon substrates using thermal processing has made it the material of choice for all modern electronics. However, as electronic devices started getting smaller and more densely packed due to the advancement of lithography technologies, finding alternatives for SiO_2 is imperative. SiO_2 faces certain disadvantages, which have resulted in a limit to the reduction of the physical sizes of the transistors, which form the building blocks of every modern electronic chip. As the thickness of SiO_2 film is reduced to below ten nanometers, it encounters issues with its leakage current, which increases due to tunneling effects. This, in turn, results in a negative impact on device performance, especially resulting in a higher power draw during operation [1-3].

This limitation of downscaling SiO_2 thin films for use in modern ultrathin transistors has resulted in researchers across the globe exploring alternate avenues. One of the most popular alternatives has been using high-*k* dielectric thin films. SiO_2 has a typical dielectric constant of ~ 4 which limits the maximum electric field that can be applied to it before its breakdown. High-*k* dielectrics, on the other hand, offer dielectric constants as high as 30. This allows them to sustain higher electric fields without breaking down. On a device level, it translates to lower leakage currents even at reduced film thickness. This allows these films to be scaled down effectively without facing the same adverse effects as the SiO_2 films. Some of the most studied high-*k* dielectrics include ZrO_2 [4-6], HfO_2 [7-9], Al_2O_3[10-12], La_2O_3[13-15] as well as other novel metal oxide-based materials [16]. In recent years, Ta_2O_5 has also been studied for its dielectric properties along with novel ways to incorporate nano structuring in the formed films [17-20]. High-*k* dielectric thin films have been deposited using several deposition techniques, such as atomic layer deposition, evaporation, sol-gel techniques, and RF magnetron sputtering [21-24]. Furthermore, techniques such as co-preciptation and galvanostatic electrodeposition could also be used for the fabrication of complex oxide films [25, 26]. However, high-*k* dielectric films do suffer from the disadvantage of a poor interface with silicon which also limits their use as a gate dielectric layer which could potentially be tackled using an interfacial layer of SiO_2 between the Si substrate and the high-k dielectric film [27-30].

A few of the critical requirements of using a dielectric film for a MOSFET include the demonstration of a high dielectric constant, low leakage current as well as a band gap offset of at least 1 eV w.r.t conduction band of silicon to avoid higher leakage currents due to tunneling [31-33]. However, typically a higher dielectric constant is accompanied by a lower band gap. In this work, we aim to tackle the issue of the trade-off between a higher dielectric constant and a lower band gap by creating a bilayer stacked structure of ZrO_2 (lower *k*, high E_g) and Ta_2O_5 (high *k*, low E_g). Furthermore, incorporating a multilayer structure also allows for larger capacitance which in turn results in easier trap filling owing to the increased density of charges induced in the semiconductor [34]. It has also been found that combining materials with individually high-performing dielectric properties could result in the formation of composites with excellent properties as well [35]. Furthermore, combining dielectric materials with appropriate polymers could result in the enhancement of the dielectric properties of the materials [36]. We study the properties of high-k dielectric thin films and their stacked structures deposited using the electron beam evaporation technique by fabricating MOSCAP devices using the dielectric films on silicon substrates. Near theoretical dielectric constant values were obtained for the high k dielectric films using SiO_2 as an intermediate interfacial layer. The resulting films demonstrate near-ideal CV curves with minimal frequency dispersion.

Experimental

SiO_2, ZrO_2, and Ta_2O_5 thin films are deposited on heavily doped N-type (001) silicon substrates and glass substrates. The substrates are cleaned using acetone, isopropyl alcohol, and DI water in an ultrasonic cleaner. The silicon substrates are then dipped in a mixture of buffered oxide etch (BOE) and hydrofluoric acid to remove the native oxide layer before the deposition. The thin films are deposited using an AJA International electron beam evaporator with an acceleration voltage of 10 kV. The thickness and the growth rate of the films are monitored using an Inficon thickness monitor. The SiO_2 thin films (~ 5 nm), ZrO_2 thin films (~ 30 nm), and the Ta_2O_5 thin films (~ 30 nm) are deposited at a growth rate of ~ 1 A^o/s with the deposition rate controlled by varying the current to the electron gun. The chamber was evacuated to a base pressure of less than 10^{-7} Torr prior to the deposition of the films. All the films are heated on a hot plate in the air at a temperature of 400 ^oC for 4 hours to improve film adhesion. Top platinum electrodes (~ 50 nm) for the MOSCAP devices were deposited using a circular shadow mask of 100 μm diameter using an AJA International DC sputtering system. Indium was used as the bottom contact for all the devices. A general representation of the fabricated MOSCAP devices is shown in Figure 1. Keithley 4200-SCS semiconductor parameter analyzer system connected to a Signatone probe station is used to characterize the fabricated MOSCAP devices to measure their IV and CV properties in ambient conditions in the dark to avoid any charge generation due to photoelectric effects. The films on the glass substrates are used to determine the band gap of the films by measuring their absorbance spectrum using a Perkin Elmer Lambda 950 UV/Vis-IR spectrometer.

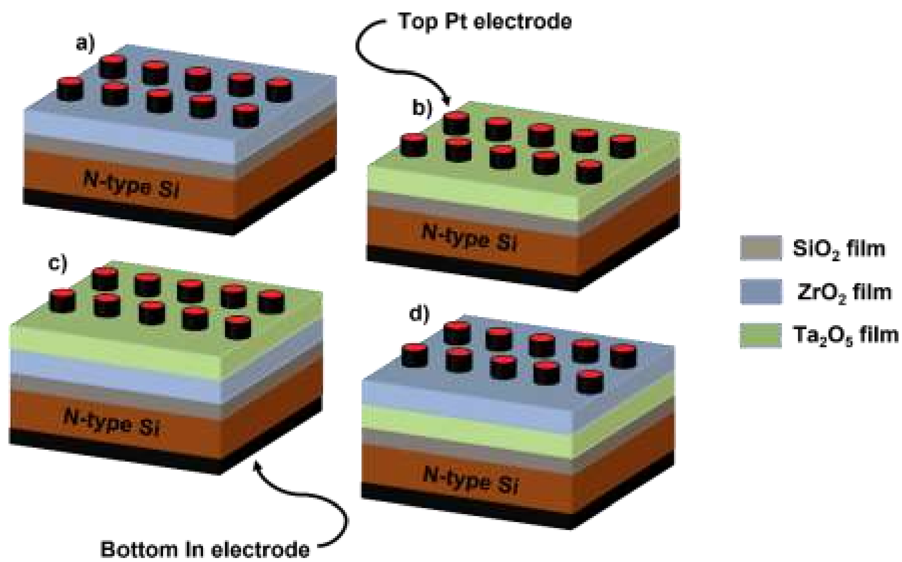

Figure 1: Schematic representation of the fabricated MOSCAP devices. (a) ZrO_2/SiO_2 (b) Ta_2O_5/SiO_2 (c) $Ta_2O_5/ZrO_2/SiO_2$ (d) $ZrO_2/Ta_2O_5/SiO_2$ are fabricated to compare the properties of the films w.r.t the stacked structures.

Results and Discussion

ZrO_2 thin films

MOSCAP devices with ZrO_2 thin film (~ 35 nm) as the dielectric are fabricated using the techniques mentioned previously. The films are characterized for their leakage current properties by measuring the current response versus the voltage. The corresponding current density curve is shown in Figure 2. The MOSCAP devices demonstrate a leakage current density value of as low as 10^{-8} A/cm^2. A lower value in the leakage current density indicates high-quality dielectric films, making them suitable for use as gate dielectrics in modern electronic devices.

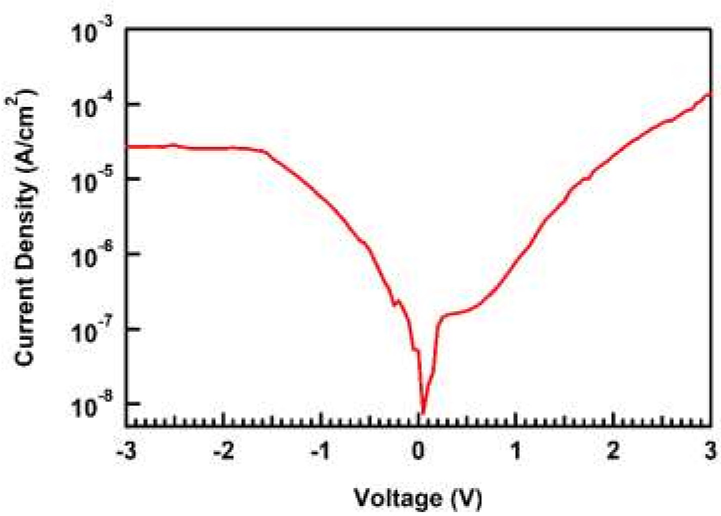

Figure 2: Current density versus voltage curve for ZrO_2 thin film-based MOSCAP devices. The curve shows a leakage current as low as 10^{-8} A/cm^2.

The optical bandgap of the films is determined using Tauc's relationship between absorbance and band gap. The absorbance of the films with varying wavelengths is determined using the UV-Vis spectroscopy measurement. The linear part of the obtained data is fitted to a straight line whose *x*-intercept determined the bandgap of the film. The absorbance spectra and the corresponding Tauc plot are shown in Figure 3 below. The band gap of the film was calculated to be ~ 4.32 eV, which was found to be lower than what was observed in the literature [37-41].

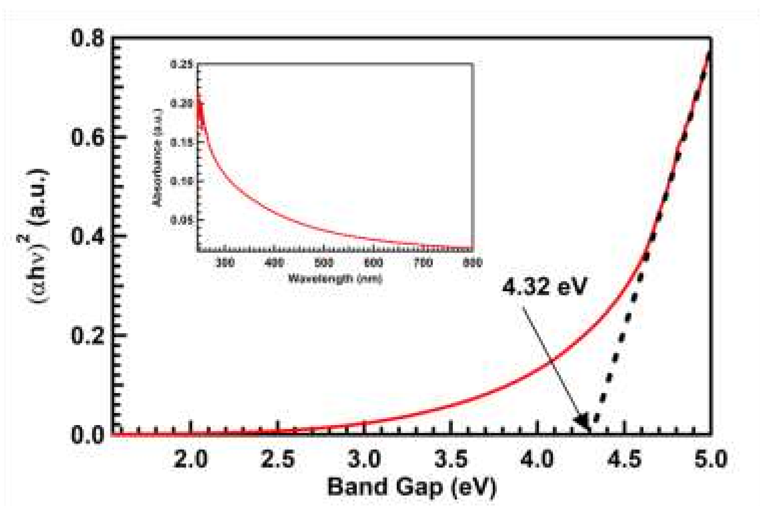

Figure 3: Absorption spectra (inset) and Tauc plot to determine the optical band gap of the ZrO_2 dielectric film. The band gap was determined to be ~ 4.32 eV.

Voltage-dependent capacitance measurements are performed in a MOSCAP configuration shown earlier (Fig 1) to determine the dielectric properties of the film. The results are shown in Figure 4 below. As seen in Figure 4(a), the films showed a clear distinction between the accumulation, inversion, and depletion regions as the voltage is swept from positive to negative voltage. A frequency dispersion is observed in CV curves as the frequency is varied between each measurement. A decrease in the capacitance value at the accumulation region was observed with increasing frequency which could be attributed to the interface state charges not being able to follow the applied AC signal at higher frequencies. The increase in the film's capacitance at lower frequencies points to the presence of charge traps on the film surface which can follow the AC signal at lower frequencies increasing the film's capacitance. Subsequently, the dielectric constant (k) of the films is calculated using the relation $k = (Cd)/(\varepsilon_0 A)$ where C is the measured capacitance, d is the film thickness, A is the area of the electrodes (in this case, circular electrodes with 100 μm diameter). The dielectric constant is plotted as a function of measurement frequency in Figure 4(b). The films demonstrated a dielectric constant greater than 30 at 1 MHz, making the film a potential candidate as a SiO_2 dielectric replacement. The observed dielectric constant was close to what has been reported earlier in the literature indicating a lack of interfacial defects due to the use of SiO_2 as an interfacial layer between the Si substrates and the high k dielectric film. A similar trend was also observed for the ZrO_2 thin film dielectrics.

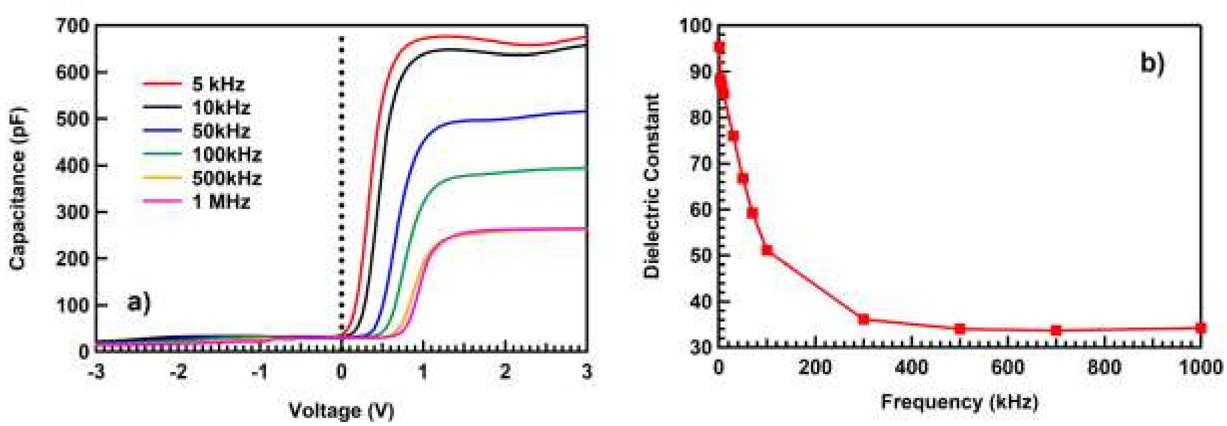

Figure 4: (a) CV curves for ZrO_2-based MOSCAP device at varied frequencies between 5 kHz and 1 MHz showing distinct accumulation, depletion, and inversion regions as well as an increased capacitance at lower frequencies. (b) Calculated dielectric constant as a function of the applied frequency.

Ta_2O_5 thin films

We fabricated 30 nm Ta_2O_5 thin films using electron beam evaporation and SiO_2 as the interfacial layer. The leakage properties of the films are determined using voltage-dependent current density measurements. The current density of the films (A/cm^2) as a function of voltage is shown in Figure 5. A leakage current of as low as 10^{-7} A/cm^2 is observed indicating a good quality film and possible use as a dielectric film.

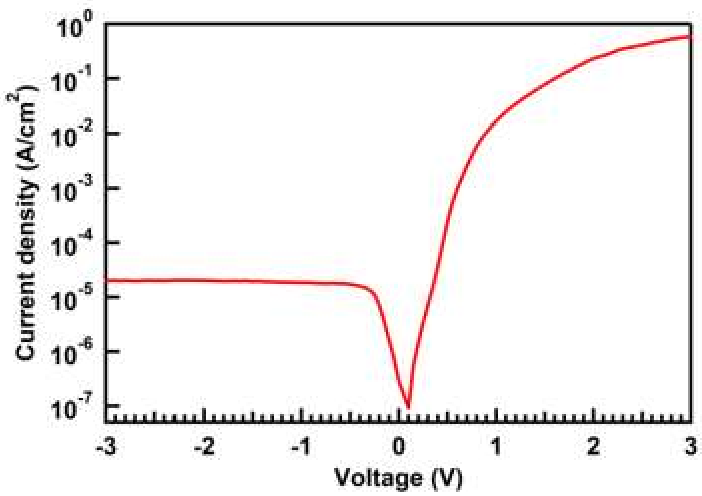

Figure 5: Voltage-dependent current density measurement on Ta_2O_5 thin films measured in a MOSCAP configuration. The films displayed a leakage current as low as 10^{-7} A/cm^2.

The film's absorbance properties (inset) as well as the calculated band gap of the film, calculated from the Tauc's relation, are shown in Figure 6 below. The films are extremely transparent as observed from the absorbance curves. The films displayed a wide band gap value of ~ 4.8 eV. A stable current density versus voltage curve as well as a wide optical band gap determined from the absorbance spectrum point towards the material being a suitable high-*k* dielectric to replace the more conventional SiO_2 dielectric.

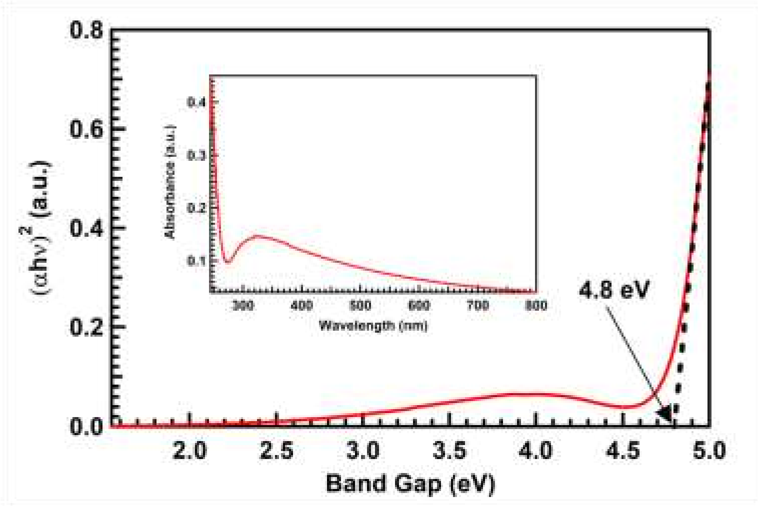

Figure 6: Absorption spectra (inset) and Tauc plot to determine the optical band gap of the Ta_2O_5 dielectric film. The band gap was determined to be ~ 4.8 eV making it a good choice to be used as a dielectric material.

Voltage and frequency-dependent capacitance measurements were performed on the films in a MOSCAP configuration to determine their dielectric properties. As seen in Figure 7, the film exhibits distinct accumulation, inversion, and depletion regions. Similar to the ZrO_2 films, the Ta_2O_5 films also displayed an increase in the capacitance values with a decrease in the measurement frequency owing to the presence of interface state charges. The dielectric constant of the film is determined to be ~ 20, which matches the value presented in other literature [42-44]. The wide band gap, low leakage current density as well as dielectric constant value make Ta_2O_5 a potential high-*k* dielectric alternative to SiO_2 films.

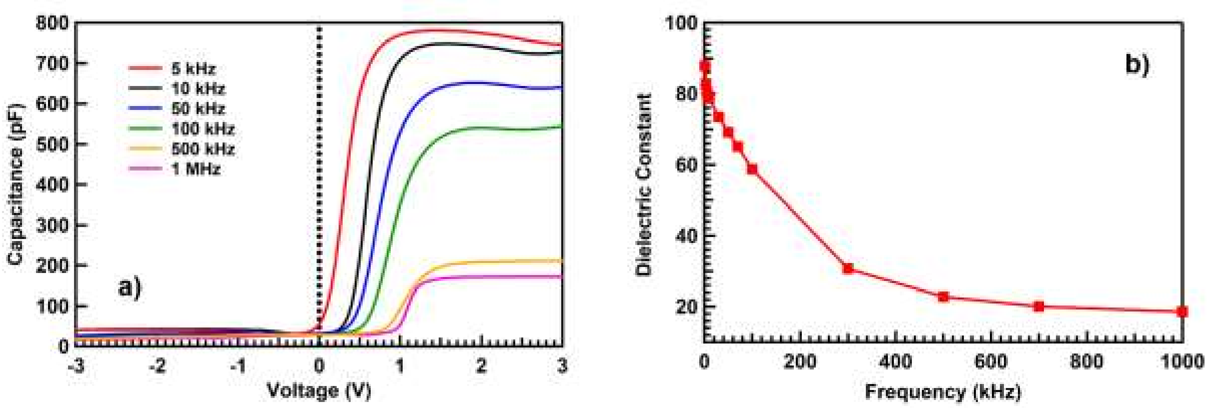

Figure 7: (a) CV measurements on Ta_2O_5 thin films demonstrating distinct accumulation, inversion, and depletion regions. (b) Calculated dielectric constant value as a function of the measurement frequency for Ta_2O_5 thin film.

Stacked dielectric films

Having demonstrated the potential of ZrO_2 and Ta_2O_5 thin films to act as a suitable SiO_2 replacement, the films are deposited in a bilayer stacked structure (Ta_2O_5/ ZrO_2 as well as ZrO_2/ Ta_2O_5) to study the effect of the high-k dielectric interface on the overall dielectric performance of the MOSCAP device. The IV performance of the fabricated stacked structures is shown in Figure 8. Both the stacked dielectric structures demonstrated a leakage current density as low as 10^{-7} A/cm^2, Figure 8(a) and 8(b). The low leakage current density observed in the films indicates the high film quality.

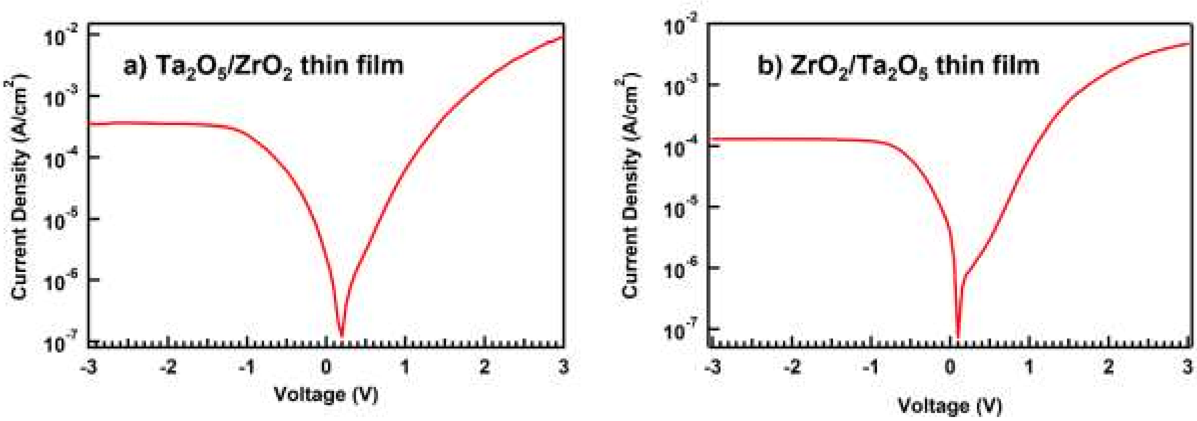

Figure 8: Leakage current density as a function of voltage for MOSCAP devices with stacked dielectrics (a) Ta_2O_5/ ZrO_2 film (b) ZrO_2/ Ta_2O_5 film.

The corresponding absorbance spectrum and the extrapolated band gap for the dielectric stacks are shown in Figures 9 and 10, respectively.

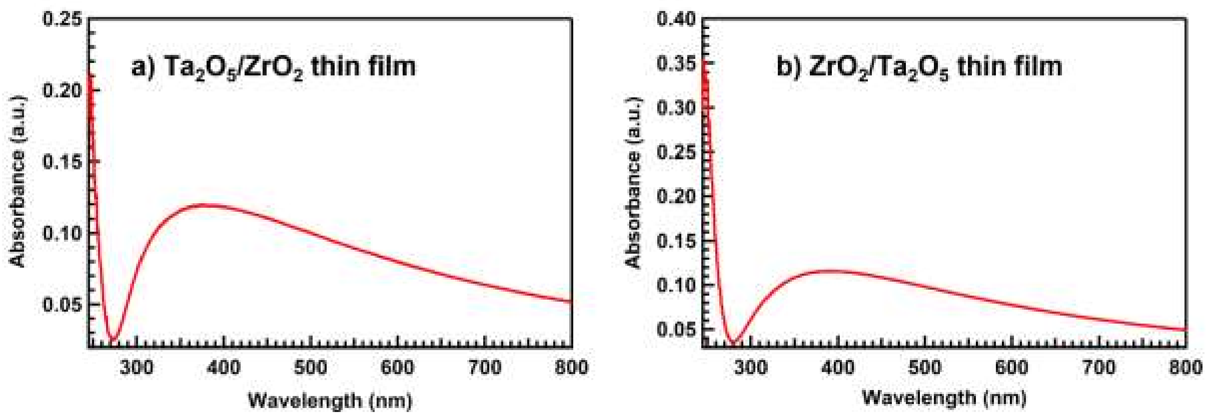

Figure 9: Absorbance spectrum for stacked dielectric films (a) Ta_2O_5/ ZrO_2 film (b) ZrO_2/ Ta_2O_5 film. The low absorbance values indicate a highly transparent film, which indicates a high band gap.

The band gaps extrapolated from the absorbance spectrum using Tauc's relation are shown in Figure 10. Both stacked dielectric structures exhibited similar bandgaps greater than 4.80 eV. The observed band gap in the dielectric stack was found to be higher than what was measured in the single-layered films by themselves. It is worth noting that the ZrO_2 thin film as a single layer demonstrated a band gap of 4.32 eV, which is considerably lower than what has been reported in the literature [31-34]. The films reported in this work are all deposited at room temperature (RT), which could result in larger particle sizes, especially using a simple evaporation technique. Furthermore, the lack of any processing gas during the deposition could also result in the presence of oxygen vacancies that could be responsible for lowering the band gap of the film [37, 45-49]. The introduction of oxygen as a processing gas and the increase in the deposition temperature and/or adding a high-temperature annealing step in the presence of an oxygen-rich atmosphere could improve the properties of the ZrO_2 thin films. This, in turn, could also be helpful in further improving the electrical properties of the stacked dielectric structures.

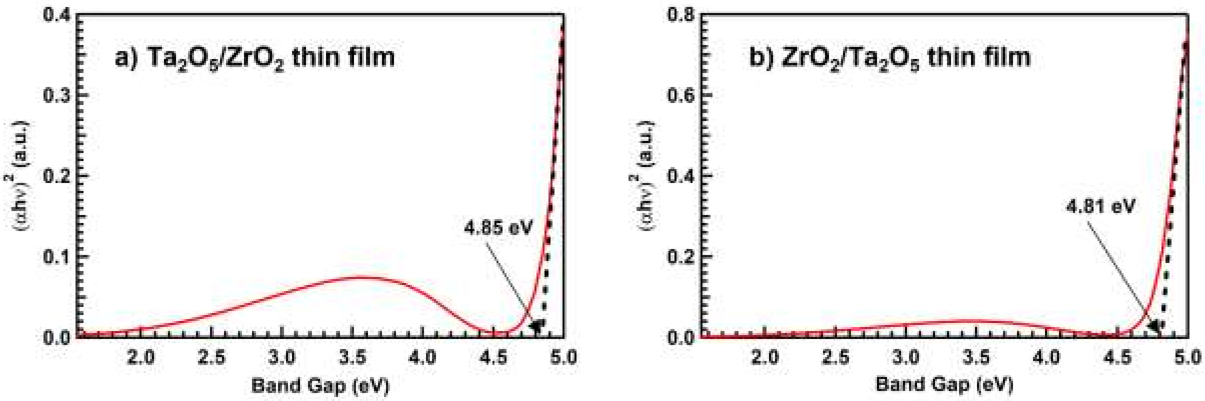

Figure 10: Extrapolated band gap of stacked dielectric from the absorbance spectra using Tauc's relation (a) Ta_2O_5/ ZrO_2 film (b) ZrO_2/ Ta_2O_5 film. Both the stacked structures showed band gaps higher than the single-layered films.

After confirming the potential of the structures to serve as high-k dielectric films based on their electrical properties, the dielectric characteristics of the films are assessed through the measurement of voltage-dependent capacitance. The CV curves are then used to determine the frequency-dependent dielectric constants of the structures using the relation used previously. The corresponding CV curves, as well as the frequency-dependent dielectric constant curves, are shown in Figures 11 and 12, respectively.

As the figures show, the CV curves exhibit distinct accumulation, inversion, and depletion regions similar to the single-layered films. An increase in the film's capacitance was observed with a decrease in frequency like the single-layered films shown previously. However, it is interesting to note that the stacked dielectric films show a lower frequency dispersion (change in capacitance versus the change in frequency) as compared to the single-layered film. Moreover, a more pronounced "bump" was observed in the CV curves of the Ta_2O_5/ ZrO_2 structure as opposed to the ZrO_2/Ta_2O_5 structure. This could be due to the ZrO_2 thin film, which was suspected to be oxygen deficient. Oxygen vacancies have significant effect on the grain size, which in turn can result in significance impact to the film properties due to the reduction of the unit cell volume [50]. This oxygen deficiency could have potentially led to surface defects at the Ta_2O_5/ ZrO_2 interface as opposed to the ZrO_2/ Ta_2O_5 interface.

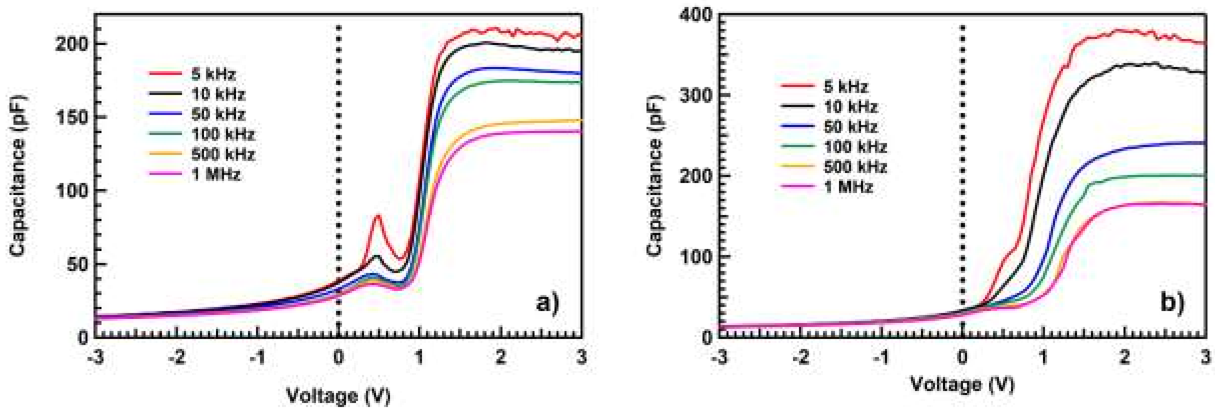

Figure 11: CV curves for stacked dielectric films in a MOSCAP configuration showing distinct accumulation, inversion, and depletion regions for (a) Ta_2O_5/ ZrO_2 film and (b) ZrO_2/ Ta_2O_5 film.

The dielectric constants of the stacked structures are determined using the relation mentioned previously. The Ta_2O_5/ ZrO_2 and the ZrO_2/ Ta_2O_5 structures showed dielectric constants of 29.3 and 32.5, respectively. The dielectric constant values are in agreement with what was observed for the single-layered films previously. One of the key advantages of these stacked structures is the improvement of the band gap of the films with minimal to no degradation in the leakage current and dielectric constant.

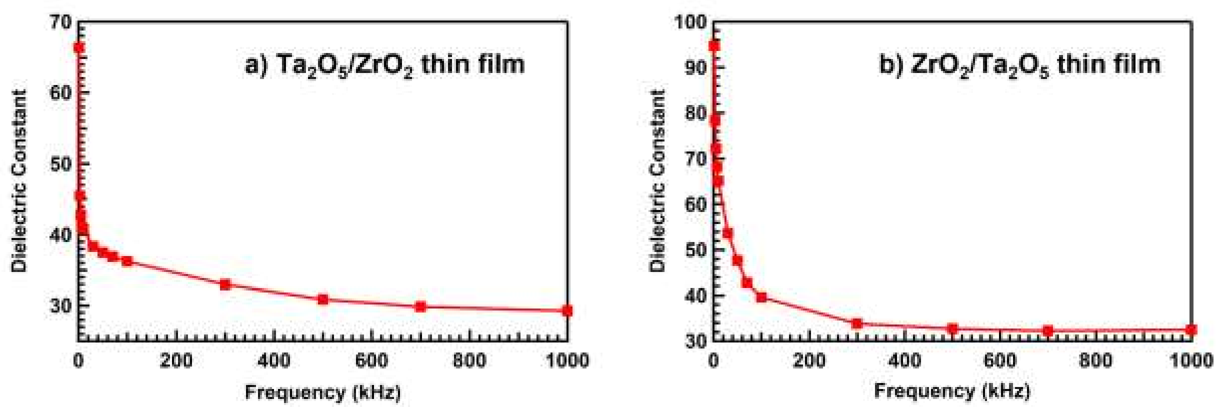

Figure 12: Calculated frequency-dependent dielectric constants for the stacked dielectric structures with (a) Ta_2O_5/ ZrO_2 film and (b) ZrO_2/ Ta_2O_5 film.

The stacked dielectric structure demonstrated dielectric constant values which are slightly lower than the single-layered films. This was also accompanied by an increase in the film's optical band gap as well as the leakage current densities. These results agree with the studies carried out by Barquinha et.al. on multilayered dielectric structures which are a mixture of high k and high band gap dielectrics [51]. The films also demonstrate dielectric constant values greater than recently studied complex oxides such as Barium and Strontium Hexaferrites [52]. However, it is to be noted that the structures fabricated in this work demonstrated a lower decrease in the dielectric constant values compared to the work shown by Barquinha et.al. It is also important to note that the dielectric films demonstrated optical bandgaps lower than the theoretical values. This could be attributed to the non-stoichiometric nature of the films, specifically related to the oxygen vacancies in the films which cause an increase in the density of states, decrease in the bandgap values as well as increase in the unit cell parameter for the films [34, 53].

Effective oxide thickness (E.O.T.) is an important characteristic of dielectric films, which helps compare the performance of a dielectric film with the more conventional SiO_2 thin film. The *EOT* of a dielectric film is determined by the relation $EOT = (k_{SiO2}/k_{high\text{-}k}) \times t_{high\text{-}k}$ where k_{SiO2}, $k_{high\text{-}k}$, and $t_{high\text{-}k}$ are the dielectric constant of SiO_2 (usually considered as 3.9), the dielectric constant of high-k dielectric film and thickness of the high-k dielectric film, respectively [37]. Typically, a lower value of *EOT* is preferred for the fabrication of MOSFETs, which indicates the possibility of obtaining dielectric performance from the high-k dielectric similar to SiO_2 film with the same thickness as the calculated *EOT* value, without having to scale down the dielectric thickness. Using the relationship above, the *EOT* of all the films was determined as shown in Table 1 along with the corresponding leakage current densities, bandgap as well as dielectric constants of the films.

It is important to note that while the stacked structures showed an *EOT* value greater than the single-layered films, the stacked structures have a physical thickness nearly double that of the single-layered films. This in turn allows the *EOT* to be reduced significantly with the reduction of the stacked structure thickness, allowing for a more controlled downscaling of devices. The low *EOT* combined with the low leakage current density, high band gap, and high dielectric constants make the stacked structure an ideal alternative solution to the more conventional SiO_2 thin film-based dielectrics.

Table 1: Summary of the electrical and dielectric properties for all the films with calculated *EOT* values. The thickness of all the films is indicated in parentheses in column 1.

Film (Thickness)	Leakage current density (A/cm^2)	Band gap (eV)	Dielectric constant @1 MHz	*E.O.T.* (nm)
ZrO_2 (36 nm)	10^{-8}	4.32	34.2	4.1
Ta2O_5 (30 nm)	10^{-7}	4.80	18.6	6.3
Ta_2O_5/ZrO_2 (58 nm)	10^{-7}	4.85	29.3	7.72
ZrO_2/Ta_2O_5 (55 nm)	10^{-7}	4.81	32.5	6.6

Conclusion

We studied the dielectric properties of ZrO_2 and Ta_2O_5 thin films and their bilayer stacked structure by fabricating MOSCAP devices using the dielectric films on silicon substrates. Near theoretical dielectric constant values were obtained for the high k dielectric films using SiO_2 as an intermediate interfacial layer. The resulting films demonstrate near-ideal CV curves with minimal frequency dispersion. The comparison between the two high k dielectric films as well as the stacked structures, while the single-layered films can act as a potential replacement to the more conventional SiO_2 dielectric, the bilayer stacked structure allows for a higher degree of control as well as improved electrical and dielectric performance. All the dielectric films are prepared using simple evaporation carried out while maintaining the substrate at room temperature. The only heating step involved in the fabrication of the MOSCAP devices is the heating of the films post-deposition to improve adhesion. This leaves the scope for future improvements, namely improving the dielectric thin films using methods such as post-annealing in an oxygen-rich atmosphere or deposition of films in the presence of oxygen as a process gas with the substrates maintained at an elevated temperature. These techniques could potentially lead to an improvement in the high k dielectric interface, in turn contributing to the overall improvement of the MOSCAP devices.

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Author Contributions

Makhes K Behera contributed to the conception, experimental design, carrying out measurements, and manuscript composition. Kelsea A Yarbrough contributed to the experimental design and carrying out measurements. Messaoud Bahoura contributed to the conception and manuscript composition.

Conflicts of interest or competing interests

The authors declare no conflicts of interest or competing interests.

Data and code availability

All the relevant data are included as part of the manuscript.

Supplementary information

Not Applicable

Ethical Approval

Not Applicable.

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