Fabricating a Dielectric Coating for an Improved Electrokinetic Micropump

S. Cenaiko[1,2], T. Lijnse[3] and C. Dalton[3,4]

[1]Chemical and Petroleum Engineering Department, University of Calgary, Calgary, Canada
[2]Centre for Bioengineering Research and Education, University of Calgary, Calgary, Canada
[3]Biomedical Engineering Graduate Program, University of Calgary, Calgary, Canada
[4]Electrical and Computer Engineering Department, University of Calgary, Calgary, Canada

Abstract— **This research proposes a method of fabricating a dielectric coating over the microelectrodes of an alternating current electrothermal (ACET) device to create a barrier between the biofluid and microelectrodes, eliminating the risk of electrolysis and creating a more effective device. Strontium titanate (STO) is proposed as a potential dielectric material for ACET devices, as it has a high dielectric constant compared to other materials, allowing for a higher device flow rate. This work examines various parameters used for the radio-frequency sputtering (rf-sputtering) of STO, and how these parameters affect the deposited film properties and the microelectrodes being coated. It was found during initial experimentation that the rf-sputtering technique used to deposit STO thin-films tends to etch away at the electrodes due to high energy oxygen ions. Literature is scarce on the topic but provides some guidance on modifications to the initial sputtering parameters. Through additional experiments, the following observations were made: a high oxygen injection (~30%) is required to ensure the STO film is nonconductive, sputtering the slides at 90° significantly reduces etching, a lower RF power reduces etching (but has not been found to eliminate it) and decreasing the bias power appears to reduce both etching and deposition rates. This work shows that a dielectric coating could be deposited over ACET electrodes with further work to optimize the parameters.**

Keywords— **strontium titanate, rf-sputtering, dielectric thin-film, AC electrothermal, electrokinetic**

I. Introduction

The development of lab-on-a-chip, organ-on-a-chip and other microdevices allows for faster, cheaper, and more efficient healthcare assessments [1]. Microdevices utilize smaller diffusion distances and large surface-to-volume ratios, enabling faster testing and monitoring of patients [2]. However, current mechanical micropumps used to manipulate fluids are not easily integrated with other microdevices and pose a notable risk to damaging fluid particulates and channels becoming clogged [3,4]. Electrokinetics includes a group of fluid manipulation techniques that utilize electric fields in place of mechanical parts to induce pumping, mixing and/or sorting of fluids and particles [5,6]. The microelectrodes used in these methods can be easily embedded within the microchannels of other microdevices using similar microfabrication techniques. In particular, the ACET technique is capable of efficiently handling high conductivity (>0.1 S/m) fluids, including biofluids (e.g., blood, urine, saliva, etc.) and pharmaceuticals [5–7].

ACET micropumps work by establishing a nonuniform AC electric field in the channel, causing the conductive solution to experience joule heating and establish a temperature gradient [7]. The resulting temperature gradient produces differences in conductivity and permittivity between fluid elements, resulting in the formation of microvortices [7]. This theory is discussed more extensively in previous work [6,8].

One of the challenges of the ACET effect is the presence of excessive electrochemical reactions at the electrodes when applying a voltage higher than about 3.5 V_{rms}, ultimately leading to device degradation and diminished pumping capacity [9,10]. A proposed way to eliminate or significantly reduce this phenomenon is to apply a non-conductive, dielectric layer over the electrodes. This would allow the electric field to act on the fluid while physically protecting the electrodes from the conductive fluid [11,12].

Typically, metal oxide dielectrics exhibit a high permittivity due to strong ionic polarization. Studies have demonstrated that metal oxides could potentially reach dielectric constants of over 120 [13-16]. A higher electric permittivity allows for minimal loss transmission of the electric field both in amplitude and power [12].

This work describes a method to fabricate the dielectric thin-films using rf-sputtering. Preliminary multiphysics simulations in COMSOL Inc. v5.5 (MA, USA) have identified STO as possessing desirable properties for ACET applications – including a high permittivity and low thermal conductivity – and is therefore investigated as a potential dielectric coating [11,12]. Various parameters are explored in this research, including the electrode material, power, bias power, substrate angle and oxygen content and pressure.

II. Materials and Methods

The fabrication of thin-films requires microfabrication techniques capable of producing thicknesses within the nanometer range, such as rf-sputtering. Microelectrodes are

generally made using photolithography, which involves sputtering a thin layer of metal on a substrate and subsequently etching away parts of the metal thin-film to pattern the electrodes [9]. The slides could then be coated with the dielectric, over an array of electrodes of an electrokinetic device. In this work, the metal layer is not patterned using photolithography, but a uniform thin-film is deposited on the slides to investigate the effects of plasma etching on the metal.

To prepare for sputtering, soda lime glass slides were first cleaned with isopropanol and acetone to remove any particles and ensure proper adhesion of STO. The slides were then sputtered with a layer of metal to produce the bottom electrode, as would be required to fabricate an ACET device; two layers of metal are used in instances where the proposed electrode material is unable to properly adhere directly to the glass. In these cases, a thin 10 nm layer of Cr or Ti metal is first deposited on the slides prior to sputtering with Au or Pt. Once the substrate was prepared, the sides of the electrode were taped off to obtain a step-height for testing the relative thicknesses of the electrode to the thin-film and the STO was sputtered over the slides. Rf-sputtering parameters and electrode materials were changed based on the results from testing. Table 1 provides the sputtering parameters for some pertinent trials. The flow rate of argon is maintained at 33 sccm for all trials, with a pressure of 10^{-3} Torr.

III. Results

Several tests were performed on each slide using a profilometer and LCR-meter to determine how sputtering parameters affect the STO deposition. The film thicknesses of slides were measured using a profilometer; the change in thickness of the electrode can be measured to determine whether the STO had deposited or if the electrode had become thinner due to plasma etching during deposition. The conductivity of the film was then tested with an LCR meter. Exact measurements could not be obtained for the resistance of the film since the thin-film tended to be nonuniformly coated, resulting in different readings at different points of the coating. Therefore, a reading of less than 20 Ω of resistance over a distance of about 1 cm was assumed to be indicative of a conductive thin-film and a reading of over 10 MΩ would indicate that the film behaves as an insulator. And finally, to test for potential charge leakages between the metal plates, some slides were sputtered with a top electrode as well to test for the resistance between the metal plates. If this reading was within a few ohms, this showed that current was easily able to pass between the electrodes through the insulative dielectric film. Table 1 includes a summary of the characteristics of the thin-films and electrodes determined through profilometry and resistance measurements.

Initial experiments, with varying oxygen content, demonstrate that the addition of oxygen to the chamber begins to show degradation of the electrodes due to negative ion bombardment [17]. The experimental setup and effects of plasma etching are depicted in Fig. 1. The oxygen ions in the plasma help to reintegrate oxygen into the STO thin-film at the substrate, but can also cause damage to previously deposited material, including the metal thin-film. The conductive nature of the STO in these trials suggests that the strontium and titanium atoms deposited on the substrate, with little to no interstitial oxygen atoms present in the film. Therefore, it is necessary to use an oxygen content of about 30% in the chamber to increase the amount of oxygen in the thin-film and produce a dielectric coating. In an effort to eliminate degradation of the electrodes, while maintaining a relatively high oxygen injection, other parameters were investigated. It was found that

Table 1 Testing parameters and results for the main trials of sputtering STO

Trial #	Electrode Material(s)	Oxygen	Power	Bias Power	Residence Time	Characteristics
1	10 nm Cr/ 100 nm Au	0%	40 W	40 W	18 h	No etching of the electrode. STO is clear and conductive.
2	10 nm Cr/ 100 nm Au	10%	40 W	40 W	18 h	The electrode is completely degraded. STO is conductive.
3	10 nm Cr/ 100 nm Au	3%	40 W	40 W	18 h	The electrode is completely degraded. STO is conductive.
4	200 nm Ti	10%	15 W	40 W	16 h	The electrode is partially degraded (~20 nm). STO is conductive.
5	N/A	30%	15 W	40 W	8 h	Sputtered at 90°. STO is nonconductive on the bottom, but conductive on the top.
6	10 nm Ti/ 100 nm Pt	30%	15 W	40 W	8 h	Sputtered at 90°. Electrode is etched only at the bottom. STO is nonconductive on the bottom, but conductive on the top.
7	10 nm Ti/ 100 nm Pt	30%	15 W	40 W	8 h	The electrode is partially degraded (~35 nm). STO is conductive. Parameters are used in accordance with reference 16.
8	10 nm Ti/ 100 nm Pt	30%	15 W	30 W	8 h	Sputtered at 90°. STO is largely nonconductive, but the bottom is conductive where the electrode was partially etched away.
9	10 nm Ti/ 100 nm Pt	30%	15 W	30 W	24 h	Sputtered at 90°. The slides were flipped 180° halfway through sputtering STO. STO is conductive.
10	10 nm Ti/ 100 nm Pt	30%	15 W	10 W	8 h	Sputtered at 90°. STO is conductive.

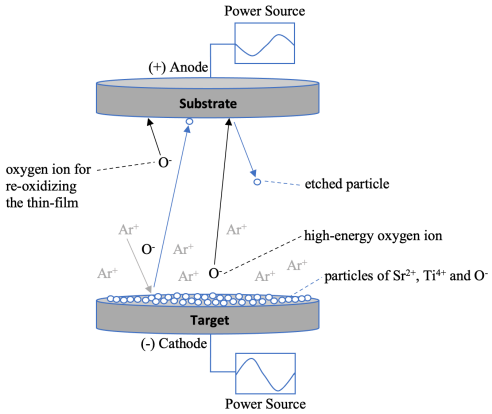

Fig. 1 Experimental setup of rf-sputtering and etching

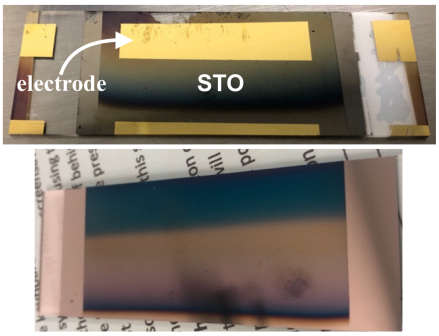

Fig. 2 Images of the results of trials #8 and #9, respectively; trial #8 is shown with a top gold electrode sputtered over the nonuniform STO coating, while trial #9 shows the changes in the colour of the STO

using Ti or Ti/Pt electrodes, instead of Cr/Au, could significantly improve the durability of the electrodes and reduced the rate of etching. One study showed that the bias power could also affect etching; it was found that as the bias power increases, so does the rate of re-sputtering, resulting in a decreased rate of deposition [18]. Experiments performed in this work are in agreement with the previous study and had similarly found that decreasing the bias power would reduce the rate of etching, but also discovered that by lowering the bias power too much the film would become conductive. It was also found that decreasing the sputtering power led to lower rates of etching. Some slides have been sputtered off-axis, at 90° to the substrate holder, as this has been suggested as another way to reduce the rate of etching of the electrodes [17]. This was found to significantly affect both the rates of etching and the electrical nature of the thin-film (whether conductive or insulative). These slides were found to be non-conductive on one side and conductive near the other edge of the slide where some etching had occurred in each case. Sputtering slides off-axis at lower angles ($\leq 30°$) were also tested, but found no significant impact on the rates of etching compared to sputtering the slides on-axis. Several hypotheses can be made with regards to why these techniques are not producing a proper dielectric thin-film and we propose methods to solve some of these issues.

IV. Discussion

These initial results demonstrate that a dielectric metal oxide can effectively be deposited over metal electrodes using rf-sputtering and paves the way for further investigation and optimization. It can be seen from these trials that the presence of oxygen in the chamber causes plasma etching to the electrodes. The ionized oxygen atoms are able to reach more than 20 times the kinetic energy of other particles in the plasma, causing negative-ion bombardment at the substrate [19]. To prevent re-sputtering, the energy with which the ions hit the substrate must be less than the bond energy of the metal and of STO [20,21]. Since platinum has a higher bond energy than gold, using a platinum electrode can increase the re-sputtering threshold energy of the electrode and reduce etching [20]. Sputtering the slides off-axis was found to reduce etching by decreasing the force with which the oxygen ions interact with the substrate, but this produces a highly nonuniform coating, as shown Fig. 2. Lowering the oxygen injection can also reduce the rate of etching, however, the oxygen is necessary for re-oxidizing the STO and producing a dielectric thin-film. The final method explored was limiting the sputtering power and bias power to reduce the kinetic energy of particles and thus reduce etching, however the bias power needed to be sufficiently high to deposit and re-oxidize the STO at the substrate.

Re-sputtering can not only damage the electrode but cause changes to the stoichiometry of the thin-film. This can result in differences in the oxygen content or atoms from the electrode becoming embedded within the dielectric, causing changes to the material properties, including the dielectric constant. Changes to the stoichiometry of the blue/purple thin-film of trial #9 (shown in Fig. 2) could be explained by this phenomenon. A potential way to more accurately determine how each of the deposition parameters discussed is affecting the deposition of STO is to use an energy-dispersive X-ray analysis (EDAX) machine to determine the elemental composition of the thin-films.

V. Conclusions

Some challenges with rf-sputtering are presented throughout this research, including degradation of the electrodes and preserving the dielectric properties of STO. The rf-sputtering of STO has been reported in literature, but typically key

parameters are not disclosed in the research articles, therefore it is necessary to further investigate these parameters [13,14]. To effectively sputter the STO thin-film, the rate of etching must be limited to ensure STO is able to form an even thin-film over the electrodes and to preserve the geometry of the electrodes for future applications in ACET devices. This study found that using a Ti/Pt electrode in place of a more common Cr/Au electrode reduces etching and thus increases the deposition rate of STO. Other ways of reducing etching at the electrodes include adopting an off-axis deposition, and lowering the sputtering and bias powers. Sputtering the slides off-axis results in an uneven thin-film, which is undesirable for ACET devices, therefore it is more practical to use other methods, such as determining the optimal sputtering and bias powers. The power sources should be sufficiently high to maximize the deposition rate and re-oxidize the dielectric thin-film, but low enough to avoid re-sputtering. Decreasing the oxygen supply was also found to lower the rate of re-sputtering, but the oxygen is also vital for maintaining the dielectric nature of STO that is required for ACET applications.

Future work includes investigating the repeatability of these results, as well as optimizing both sputtering parameters and the oxygen injection to develop an even coating of STO and maximize the dielectric constant based on the stoichiometry of the thin-film. In this work, the pressure in the chamber was constant throughout the experiments, but some research suggests that further increasing the pressure can decrease the effects of negative ion bombardment [17]. Other potential ideas to explore include using a different dielectric material that is more commonly used and easier to work with, such as aluminium oxide, or using a different method for depositing STO that may have less issues interacting with the electrodes, such as chemical vapour deposition (CVD).

Acknowledgment

We would like to acknowledge Dylan Finch and Malcolm Haynes of the University of Calgary's Microsystems Hub cleanroom facility, for assisting with equipment and experiments, and NSERC for financial assistance.

Conflict of Interest

The authors declare that they have no conflict of interest.

References

1. Mitra D, Ghoshal S, Rahaman H et al (2011) Test Planning in Digital Microfluidic Biochips Using Efficient Eulerization Techniques. J Electron Test 27(5):657–671 DOI 10.1007/s10836-011-5239-2
2. Lai S, Wang S, Luo J et al (2004) Design of a Compact Disk-like Microfluidic Platform for Enzyme-Linked Immunosorbent Assay. Anal Chem 76:1832–1837 DOI 10.1021/ac0348322
3. Sutera S P (1977) Flow-Induced Trauma to Blood Cells. Circ Res 41:2-8 DOI 10.1161/01.RES.41.1.2
4. Amirouche F, Zhou Y, Johnson T (2009) Current micropump technologies and their biomedical applications. Microsyst Technol 15(5):647–666 DOI 10.1007/s00542-009-0804-7
5. Lian M, Islam N, Wu J (2007) AC electrothermal manipulation of conductive fluids and particles for lab-chip applications. IET Nanobiotechnol 1(3):36–43 DOI 10.1049/iet-nbt:20060022
6. Salari A, Navi M, Lijnse T et al (2019) AC Electrothermal Effect in Microfluidics: A Review. Micromachines (Basel) 10(11):762 DOI 10.3390/mi10110762
7. Lang Q, Wu Y, Ren Y et al (2015) AC Electrothermal Circulatory Pumping Chip for Cell Culture. ACS Appl Mater Interfaces 7(48):26792–26801 DOI 10.1021/acsami.5b08863
8. Ramos A, Morgan H, Green N G et al (1998) Ac Electrokinetics: a Review of Forces in Microelectrode Structures. J Phys 31:2338–2353 DOI 10.1088/0022-3727/31/18/021
9. Vafaie R H, Ghavifekr H B, Van Lintel H et al (2016) Bi-directional ACET micropump for on-chip biological applications. Electrophoresis 37:719-726 DOI 10.1002/elps.201500404
10. Salari A, Navi M, Dalton C (2014) IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), Des Moines, USA, 2014, pp 1–4
11. Lijnse T, Cenaiko S, Dalton C (2019) Prevention of Electrode Degradation in ACET Micropumps for Biomedical Devices, Alberta Biomedical Engineering Conference, Banff, Canada, 2019
12. Lijnse T, Cenaiko S, Dalton C (2020) Numerical simulation of a tuneable reversible flow design for practical ACET devices. SN Appl Sci 2:305 DOI 10.1007/s42452-020-2098-4
13. Wang Z, Kugler V, Helmersson U et al (2002) Characteristics of SrTiO3 Thin Films Deposited on Si by Rf Magnetron Sputtering at Various Substrate Temperatures. Philos Mag 82:891–903 DOI 10.1080/13642810208218351
14. Baumert B A, Chang L H, Matsuda A T et al (1997) Characterization of Sputtered Barium Strontium Titanate and Strontium Titanate-Thin Films. J Appl Phys 82:2558–2566 DOI 10.1063/1.366066
15. Stewart D M, Lad R J (2018) Enhanced Crystallinity of h-BN Films Induced by Substrate Bias During Magnetron Sputtering. Phys Status Solidi B 255:1700458 DOI 10.1002/pssb.201700458
16. Reynolds G J, Kratzer M, Dubs M et al (2012) Sputtered Modified Barium Titanate for Thin-Film Capacitor Applications. Materials, 5:575–589 DOI 10.3390/ma5040575
17. Hadj Y A, Ambriz V F, Amaechi I et al (2018) Impact of negative oxygen ions on the deposition processes of RF-magnetron sputtered SrTiO3 thin films. Thin Solid Films 661:23–31 DOI 10.1016/j.tsf.2018.05.054
18. Martin N, Baretti D, Rousselot C et al (1998) The effect of bias power on some properties of titanium and titanium oxide films prepared by r.f. magnetron sputtering. Surf Coat Technol 107(2-3):172–182 DOI 10.1016/s0257-8972(98)00647-1
19. Poulain R, Proost J, Klein A (2019) Sputter Deposition of Transition Metal Oxides on Silicon: Evidencing the Role of Oxygen Bombardment for Fermi-Level Pinning. Phys Status Solidi A 216(23):1900730 DOI 10.1002/pssa.201900730
20. Yan C, Zhang Q Y (2012) Study on low-energy sputtering near the threshold energy by molecular dynamics simulations. AIP Adv 2(3):32107–32115 DOI 10.1063/1.4738951
21. Evstifeev V V, Kostina N V (2020) Numerical Study of the Thresholds of Sputtering of Metals during Bombardment with Low-Energy Ions. Bull Russ Acad Sci 84(6):720–726 DOI 10.3103/S1062873820060118