Vacuum 114 (2015) 49-53

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Aerosol deposition-based micropatterning of barium titanate via sulphur hexafluoride inductively coupled plasma etching



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ARTICLE INFO

Article history: Received 20 July 2014 Received in revised form 21 December 2014 Accepted 23 December 2014 Available online 8 January 2015

Keywords: Micropattern formation Aerosol deposition Barium titanate Sulphur hexafluoride Inductively coupled plasma etching

ABSTRACT

Aerosol deposition (AD) is an advantageous alternative to the conventional thin film deposition method and thermal spray coating technology; this process is a very attractive method for micropatterning barium titanate (BTO) materials. AD features a high deposition rate, low process temperatures, fine patterning for miniaturisation, and improved device performance for many applications, such as RF components, dynamic random access memory, micro-electro-mechanical systems, and optical devices. In this study, BTO micropatterns deposited by AD are combined with inductively coupled plasma (ICP) etching technology in SF₆/O₂/Ar plasmas to yield a lateral resolution reaching approximately 500 nm, which is at least 10 times smaller than the resolutions achieved by other recent deposition technologies. The experimental results indicate that adding the appropriate quantities of O₂ and Ar to SF₆ can improve the etching properties by increasing the etching rate of BTO films while decreasing the surface roughness of the Pt layers. The maximum etching rate is 67.5 nm/min when the SF₆/O₂/Ar composition is 75/5/ 10 sccm, which is 15 times faster than the conventional plasma etching process. In this study, the micropatterning of AD-deposited BTO films is systematically investigated and analysed while using sulphur hexafluoride ICP etching.

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1. Introduction

Currently, complementary metal-oxide-semiconductor (CMOS) scaling faces a fundamental barrier because a ferroelectric layer must be used as a gate oxide to reduce the sub-threshold slope [1] and to boost the on-current behaviour [2]. These enhancements occur because ferroelectric negative capacitance results after the "Boltzmann limit" of the transistor power dissipation are overcome [3]. Barium titanate (BTO) thin films are excellent candidates for use in these applications due to their high dielectric constant, low leakage current, and good ferroelectric properties among numerous ferroelectrics that have been applied for low-power nanoscale applications [4,5]. Several deposition technologies have been developed to prepare BTO films, such as the sol–gel method [6], RF sputtering [7], and molecular beam epitaxy [8]. However, these methods require complicated fabrication processes and induce high costs. In this article, we focus on the aerosol deposition

(AD) process because it is a low-temperature and low-cost method featuring room-temperature processing, a high deposition rate, and a high density [9–11].

Integrating ferroelectrics into silicon-related manufacturing processes is a major technological challenge. Micropatterns must be developed with features that are as small as possible when producing highly integrated metal-oxide-semiconductor field effect transistors (MOSFETs) involving BTO thin films. Therefore, the development of a suitable anisotropic etching process for BTO thin films is important when attempting to obtain small feature sizes, accurate pattern transfers, and smooth surface morphologies. The majority of the work conducted to understand the etching mechanisms has focused on elucidating the optimal gas chemistry and process conditions necessary for achieving a high etching rate and a smooth surface morphology.

In this work, the BTO etching rate and Pt surface roughness were investigated relative to the SF₆/O₂/Ar mixing ratio by observing the etching characteristics of BTO films in SF₆/O₂/Ar plasma. Consequently, a maximum etching rate of 67.5 nm/min and a minimum micropatterning width of 0.5 μ m for 300-nm-thick BTO thin films were obtained.





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2. Experimental procedure

The BTO thin films were deposited via an AD process using a commercial crystalline BaTiO₃ powders (SBT-03B, Samsung Fine Chemicals Co., Ltd., Ulsan, South Korea) with the average particle size of 300 nm and high density over 95% of the bulk density [12] on 4-inch Pt/Ti/SiO₂/silicon substrates. The particles are aerosolised in an aerosol chamber and transported into a deposition chamber using 5 L/min N₂ gas. The transported BTO powders were continuously ejected through the nozzle and deposited onto the silicon substrate. The orifice size of the nozzle, the deposition area, the distance between the nozzle and the substrate, the working pressure, and the deposition time were $10 \times 0.4 \text{ mm}^2$ (10 mm wide, 0.4 mm slit width), 10×10 mm², 5 mm, 3.4 Torr, and 10 min, respectively. The final thickness of each BTO film was approximately 300 nm. A negative photoresist, specifically, 3.5µm-thick DNR-L300-40 (Dongjin Semichem Co., Ltd., Seoul, South Korea), was spun at 5000 rpm for 40 s in the track before being baked for 90 s at 90 °C. Afterwards, a photolithographic process was performed with an exposure energy of 120 mJ/cm² and a post-exposure bake (PEB) was performed for 90 s at 100 °C. The photoresist was then developed in an AZ300MIF developer (AZ Electronic Materials USA Corp., NJ, USA) for 60 s. Next, a 10/790nm Ti/Cr metal shadow mask was fabricated by e-beam evaporation and used during the BTO etching. After the metallisation process, the photoresist was stripped using acetone. The BTO films were etched in an ICP etching system (STS Multiplex ICP ASE Etcher). The flow rates of SF₆, O₂, and Ar gases into the operation chamber were controlled by mass flow controllers. The BTO films were etched under the following conditions: the flow rates of SF₆/ O₂/Ar were 50/0/10, 50/2.5/10, 50/5/10, 50/7.5/10, 50/10/10, 50/5/ 0, 50/5/5, 50/5/15, 50/5/20, 25/5/10, 75/5/10, and 100/5/10 sccm. Etching process was performed under fixed parameters: a bias power of 150 W, a source power of 1000 W, a total gas pressure of 7.5 mTorr, and a chamber temperature of 293 K. Finally, the Ti/Cr shadow mask was stripped using hydrofluoric acid and a Cr etchant; the etched BTO films were post-annealed at 750 °C for 1 min under N₂ atmosphere via rapid thermal annealing to completely remove all of the fluoride formed during the etching process.

The different samples have been tested repeatedly by varying the etching gas flow rates in terms of BTO etching rate and Pt surface morphology and the outcomes of each test has been individually analysed before proposing the final optimised outcome. The etching rates of the BTO films and the Ti/Cr shadow mask were verified using a focused ion beam (FIB) under stationary etching conditions. The surface morphology of the lower Pt layer was examined using atomic force microscopy (AFM). Finally, the top and cross-sectional views of the micropatterned BTO films were obtained using scanning electron microscopy (SEM) and FIB, respectively. The experimental work-flow of the proposed AD-based BTO micropatterning process is shown in Fig. 1.

3. Discussion and results

The performance of $SF_6/O_2/Ar$ plasma must be understood because the etching process is affected by changes in the chemically reactive radicals and ions on the etching surface of the BTO films [13]. The major chemical reactions potentially involved in the BTO etching process are as follows:



Fig. 1. Schematic diagram of the AD-based BTO micropatterning process.

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