Journal of Alloys and Compounds 690 (2017) 249–255

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Dielectric properties under high electric field for silicon doped alumina thin film with glass-like structure derived from sol-gel process

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A R T I C L E I N F O

Article history:
Received 15 April 2016
Received in revised form
4 July 2016
Accepted 12 July 2016
Available online 21 August 2016

Keywords:
Alumina thin films
Si doping
Glass-like structure
Breakdown strength
Leakage current
Dielectric constant and loss

A B S T R A C T

Dielectric $Al_{2-x}Si_xO_y$ (X = 0.00, 0.02, 0.05, 0.10) thin films were deposited onto $Pt/Ti/SiO_2/Si$ substrates using sol-gel spin coating technology. The obtained materials were characterized via differential scanning calorimetry (DSC), scanning electron microscopy (SEM), fourier transform infrared spectrometry (FT-IR) and X-ray photoelectron spectrometry (XPS). The results show that the films are amorphous with Si atoms occupying Al atom sites forming Al—*O*—Si bonds and glass-like structure. The dielectric properties of the film were investigated. By means of silicon doping, the leakage current and the dielectric loss of the amorphous alumina films much reduced while the breakdown strength enhanced. Two orders of magnitude reduction in leakage current and significant enhancement in breakdown strength (up to 566 MV/m) can be achieved. The improved dielectric properties are attributed to the forming of Al—*O*—Si bonds and cation vacancies by the Si-addition. The structure modification enhanced the stability of alumina structure and promoted the ionic transportation to repair the defects of the alumina films.

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1. Introduction

In the recent decade, the development of green energy technology promoted extensive research effort on energy storage technology. Dielectric capacitors as a kind traditional energy storage device are still highly important for high density energy storage applications [1–3]. For dielectric energy storage applications, high dielectric constant and high breakdown strength are highly desirable. Polymer and polymer composite with very high electric breakdown strength are widely used in capacitor industry to develop high voltage and high energy storage capacitors [4,5]. Inorganic ceramic and glass ceramic dielectrics are another alternative perspective way to develop high performance energy storage devices.

Alumina (Al_2O_3), as an excellent insulating dielectric material, is widely used as substrate and packaging material in electronics and microelectronics [6]. Moreover, because of its high breakdown strength (300–700 MV/m), high relative permittivity (8.6–10) and high band gap (~9 eV) in conjunction with its excellent chemical and thermal stability, more and more attentions has been drawn to explore its potential applications in high energy density storage capacitors [7].

Alumina thin film can be prepared via conventional sol-gel and spin coating route. Dense, homogeneous and uniform film can be prepared at low temperatures [8]. One of the distinct advantages of the sol-gel technology is: the chemical composition and phase composition of the material can be easily and precisely controlled. Recently, the substitution effect of doping elements (Ti^{4+}, La^{3+}) on the leakage current and breakdown characteristic of amorphous alumina thin films has been studied and excellent dielectric properties of doped alumina thin films were achieved [9,10]. Silicon is one of the very common elements in glasses and ceramics. Typically, Silicate glass has a high hardness and mechanical strength owing to the forming of a stable amorphous Si—O network structure [11,12]. It is interesting to see if silicon can be introduced into the amorphous alumina thin film to enhance the stability and dielectric behavior of the alumina structure. The current work is mainly focused on the dielectric properties of Si-doped alumina thin film under high electric field.

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http://dx.doi.org/10.1016/j.jallcom.2016.07.125
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2. Experimental

2.1. Sample preparation

The $Al_{2-x}Si_xO_Y$ (x = 0, 0.02, 0.05 and 0.1) thin films represented as Si-0%, Si-2%, Si-5%, Si-10% respectively, were prepared by the sol-gel and spin coating technology. Aluminum isopropoxide ($Al(OC_3H_7)_3$) and tetraethyl orthosilioate ($C_8H_{20}O_2Si$) were used as Al-precursor and Si dopant, respectively. Firstly, both aluminum isopropoxide and tetraethyl orthosilioate were dissolved in glycol ether under constant stirring at 60 °C for 30 min. Secondly, 0.02 mol acetylacetone acting as chelating agent was added to restrain the hydrolysis of Aluminum isopropoxide with stirring for 30 min. The mixture was then heated to 90 °C and 10 ml acetic acid as catalyst was added to the above solution with agitation for 30 min. Finally, the mixture was cooled down to room temperature to get a clear and homogeneous sol. Prior to spin coating deposition of the film, substrates ($Pt/Ti/SiO_2/Si$) were ultrasonically rinsed in acetone, deionized water and ethyl alcohol, successively, and dried by blowing nitrogen N_2.

Thin films were deposited on the substrate by spin-coating method at 3000 rpm for 20 s per each layer in a clean-room environment. After each deposition, the films were preheated in a tubular furnace at 450 °C for 5 min to form solid films through evaporating the solvents and burning-out organic residuals. The above procedure was repeated for 7 times to obtain samples. Samples were annealed at 600 °C for 3 h with a heating speed of 3 °C/min and then cooled down to room temperature in a muffle furnace. Gold was deposited on to the alumina film using a vacuum evaporation instrument (ZHD-400, Technol Science, China) via a mask as top electrode to form Metal-Insulator-Metal (MIM) structure with Pt as bottom electrode for the measurement of the dielectric properties. The diameter of the Au top electrode is 1 mm.

2.2. Characterizations

Surface morphology of Si-doped alumina thin films was characterized by field emission scanning electron microscopy (FESEM) (S-4700, Hitachi, Japan). The thickness was measured by a dual beam laser interferometer (F20 Filmetrics Inc. San Diego, US). The existence form of silicon in Si-doped alumina thin films was analyzed by fourier transform infrared spectroscopy (FT-IR) (EQUINOX 55, Bruker Optics, Germany) and X-ray photoelectron spectroscopy (XPS) (ESCALAB 250Xi, Thermo Fisher, USA). The combustion temperature of organics and crystallization of the samples was recorded by differential scanning calorimeter (DSC) (SAT449C, Netzsch, Germany) from 30 °C to 1200 °C at a heating rate of 10 °C/min in nitrogen atmosphere. Dielectric behaviors of the alumina film samples were measured. Before the measurements, samples were treated at 100 °C for 30 min to remove any adsorbed water. The leakage current as a function of voltage was measured using a Keithley 2400 source meter unit interfaced with a computer to perform the measurements and record the experimental data. Voltage was applied in a successive voltage step of 0.2 V/step with a delay time of 0.1 s/step, until the leakage current increased sharply and abruptly, indicating the occurrence of breakdown. The positive voltage was applied to the Au top electrode, and the Pt bottom electrode was grounded. For each sample, 10 breakdown trials were tested to estimate the breakdown strength through Weibull distribution function due to the randomness of the dielectric breakdown voltage. The dielectric constant and loss of the alumina film were measured from 100 Hz to 2 MHz using a LCR meter (E4980A, Agilent, USA).

3. Results and discussion

3.1. DSC analysis of samples

The thermal decomposition and transformation processes of the Si-0%, Si-2%, Si-5% and Si-10% samples at a heating rate of 10 °C/min in nitrogen (99.99% purity) atmosphere are described by the differential scanning calorimetry curves as shown in Fig. 1. The thermal decomposition and transformation processes of the samples can be divided into three different stages. During the initial stage (S-1), when the temperature is low (30–320 °C), there are two endothermic peaks at around 85 °C and less than 300 °C (214 °C for

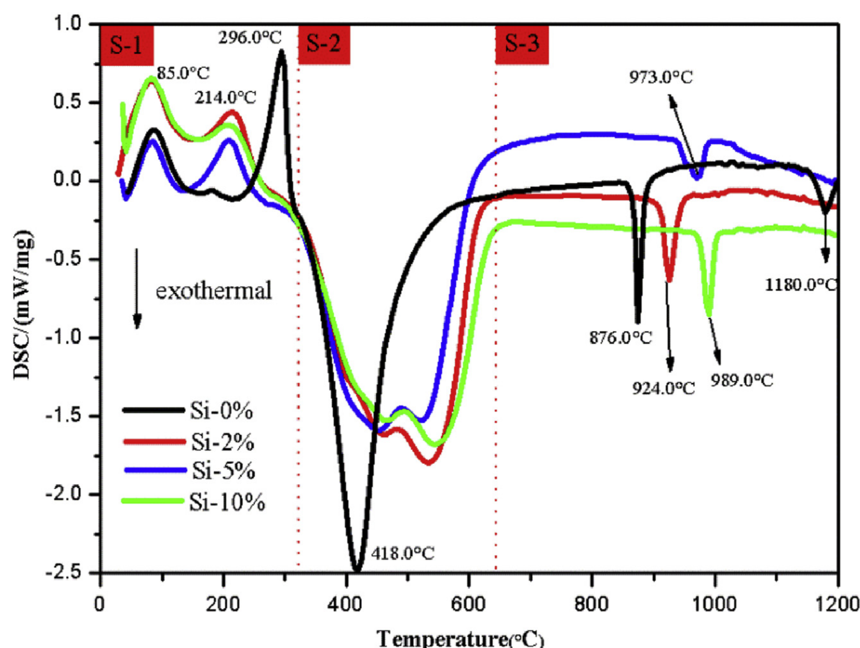

Fig. 1. DSC curves of Si-0%, Si-2%, Si-5% and Si-10% at 10 °C/min.

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