Thin Solid Films 546 (2013) 22–25

Contents lists available at ScienceDirect

Thin Solid Films

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

Effect of substrate temperature on the properties of electron beam deposited tantalum films

Nishat Arshi, Junqing Lu *, Chan Gyu Lee, Bon Heun Koo, Faheem Ahmed

School of Nano & Advanced Materials Engineering, Changwon National University, Changwon, Gyeongnam 641-773, Republic of Korea

ARTICLE INFO

Available online 16 May 2013

Keywords:
Tantalum
Thin films
e-Beam evaporation
Resistivity
XRD

ABSTRACT

In this work, the effect of substrate temperature (room temperature — 250 °C) on the structural, morphological and electrical properties of tantalum films deposited on Si/SiO_2 (100) substrate by electron beam evaporation technique was presented. The structural analysis of the deposited films was done using X-ray diffraction (XRD). The XRD patterns revealed the growth of tetragonal crystalline structure (β-Ta) and that the crystallinity of the films increased with the increase of substrate temperature. The field emission scanning electron microscopy (FESEM) images showed that the film morphology was smooth with fine spherical particles on the surface. The film thickness measured using a cross-sectional FESEM was found to increase from 97 nm to 165 nm with the increase in substrate temperature. The resistivity of tantalum films was found to decrease from ~350 μΩ-cm to ~220 μΩ-cm with increasing substrate temperature from room temperature — 250 °C respectively.

1. Introduction

Tantalum films have generated significant research interests due to their potential applications for mechanical and microelectronic industry [1–5]. Tantalum films have many attractive characteristics like high melting temperature, high ductility, good wear and corrosion-resistance, chemical inertness and temperature stable electrical resistivity [1–5]. It is one of the most promising potential diffusion barrier materials for copper metallization in integrated circuit devices [6]. Mainly, two crystalline phases of tantalum film can be formed in a deposited film: one is α-phase tantalum (body centered cubic (bcc)) with low resistivity of 15–60 μΩ-cm and the other is β-phase tantalum (tetragonal) with high resistivity of 170–210 μΩ-cm. These phases differ in both structural and electrical properties [7]. The α-Ta is considered as an attractive coating material in many applications for its high toughness, ductility, low electrical resistivity (15–60 μΩ-cm), and corrosion resistance [8]. The β phase of tantalum has been attracting much interest since its discovery in 1965 by Read and Altman [9]. Because of its high resistivity (170–210 μΩ-cm) [8], it is the preferred form for fabricating resistors and capacitors. The coexistence of α and β phases in thin films limited their use in microelectronics industry. However, the preparation of tantalum films seems to be of continuing interest. Beside these two crystalline phases, tantalum films can also posses amorphous structure. The main difference between these phases is the large difference in their electrical resistivities, i.e. >250 μΩ cm.

Various reports have been focused on developing deposition techniques and finding appropriate deposition conditions for obtaining alpha (α) and/or beta (β) tantalum films, mainly because of its varied properties and variety of applications [10]. The properties and crystal structures of tantalum films are dependent on the deposition technique, deposition pressure, substrate material, temperature, and also the geometry of the deposition system [11,12]. The crystallization orientation and grain size are amongst the most important microstructure characteristics that significantly affect the electrical properties of the films. Various methods have been employed to deposit and study the properties of tantalum films using different sputtering techniques, such as DC sputtering [13,14], RF sputtering [14], triode sputtering [15,16], ion beam assisted sputtering [17], and magnetron sputtering [18,19]. Chemical vapor deposition [20] and electrochemical deposition [21] have also been used. But, only a few reports are present for depositing and studying the properties of tantalum films using electron beam (e-beam) evaporation technique [22,23]. The formation of phases in a tantalum film depends on the deposition conditions; for example deposition pressure, substrate temperature and film thickness [11,12]. However, the phase of tantalum films is usually affected by substrate temperature. The structure of tantalum films at different substrate temperatures can be bcc α-phase, β-phase or a mixture of the two phases. The β-Ta phase of tantalum is predominant in thin films deposited at low temperature and is known to be thermally unstable and transforms to the bcc phase at temperatures above 750 °C [5,24]. However, the mechanism of the preferential growth of β-Ta phase is not well understood and a broad range of resistivity has been reported [8]. Previous work has focused on determining the experimental conditions of thin films for which α and/or

* Corresponding author. Tel.: +82 55 213 3698; fax: +82 55 261 7017.
E-mail address: junqinglu@yahoo.com (J. Lu).

0040-6090/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.tsf.2013.05.026

β tantalum form. The interesting and sometimes confusing point concerning tantalum films is the allotropy. With proper choice of deposition conditions and substrate temperature that does not compromise the film properties, tetragonal phase tantalum films with excellent functional properties can be deposited by e-beam evaporation technique.

In this work, the growth of pure β-phase tantalum films has been accomplished by low substrate temperatures (room temperature — 250 °C) that do not deteriorate the film properties and make them potential for diffusion barrier applications. In particular, we present the results of investigation on the effects of substrate temperature (room temperature to 250 °C) on the structural, morphological and electrical properties of the tantalum films deposited by e-beam evaporation technique on Si/SiO_2 (100) substrates. Until now, the systematic study on the effect of this low temperature deposition range by e-beam method has not been reported. Our demonstration of the ability to produce a fully tetragonal-phase tantalum films at substrate temperature ranging from room temperature — 250 °C using e-beam brings the required conditions closer to the realm of practical diffusion barrier applications. This study will provide a bridge between laboratories and industries to increase the utilization of tantalum films as a Cu diffusion barrier.

2. Experimental details

The tantalum films were deposited by e-beam evaporation technique onto Si/SiO_2 (100) substrate by evaporating tantalum metal. The films were deposited on Si/SiO_2 (100) substrates (area = 40×20 mm^2), which were oxidized in a furnace for one hour at 1000 °C and cleaned by rinsing in ultrasonic baths of acetone and methanol prior to the deposition. Commercial tantalum metal (99.9999%; 3.2 mm diameter × 3.2 mm length, purchased from Alfa Aesar) slugs were placed into a crucible as evaporation sources for film deposition. The vacuum chamber was equipped with a turbo-molecular pump, which is horizontally fixed to the chamber, backed by a rotary pump and could produce an ultimate vacuum of 2.8×10^{-4} Pa. A series of films were deposited for various substrate temperatures (room temperature, 50 °C, 100 °C, 150 °C, 200 °C and 250 °C) for 20 min.

The crystallographic structure of the films were analyzed by means of X-ray diffraction using (Phillips X'pert; MPD 3040) X–ray powder diffractometer with Cu Kα radiations (λ = 1.5406 Å) operated at 40 kV and 30 mA. The XRD pattern was recorded within the scan range of 30 to 60° and step size of 0.02°. The morphology and the thickness of the film were characterized by field emission scanning electron microscopy (FESEM) using a TESCAN; MIRA II LMH microscope operated at 15 kV. The elemental composition was determined by energy dispersive X-ray spectroscopy (EDX, Inca Oxford, attached to the FE-SEM) with an operating voltage of 15 kV within the energy range of 0–10 keV. The electrical resistivity of the deposited tantalum films was measured at room temperature by four point probe (Keithley — 2002) method.

3. Results and discussion

The XRD patterns of tantalum films grown at different substrate temperatures are shown in Fig. 1. All the samples deposited with different substrate temperatures on Si/SiO_2 (100) substrate show single peak, with 2θ position at 37.39° identified as (330) peak for the β phase tetragonal crystalline structure of tantalum. These results are in good agreement with the recently reported work by Dorranian et al. [25]. However, the present results are in contrast with many earlier reports, which showed multiple peaks for tantalum [5,26]. In the present work, on increasing the substrate temperature the intensity of the peak corresponding to Ta (330) plane is increased with a slight decrease in peak width and no change in phase from β-Ta to α-Ta was observed. This increase in the peak intensity with increase

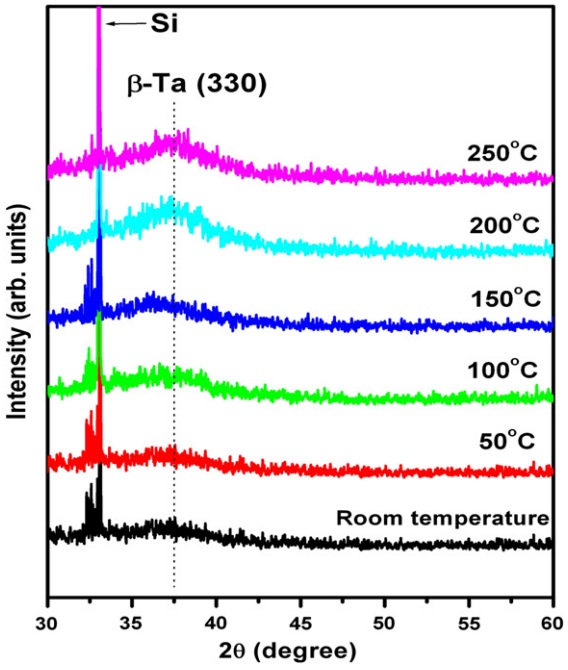

Fig. 1. XRD Patterns of the tantalum films deposited on Si/SiO_2 substrates at different substrate temperature.

in substrate temperature indicates an improvement in the crystalline nature of the film [27]. In general, the substrate temperature usually affects the growth of films by increasing the surface mobility of the arriving atoms. The condensation of tantalum atoms on the cold substrates results in the formation of disordered metastable structures [25]. Additionally, the grain size of the tantalum films was also found to be dependent on the substrate temperature. The decrease in the width of the peak at high substrate temperature, interpreted according to the formula of Scherrer [28] corresponds to an increase in the grain size from 20 to 35 nm. These results are also in good agreement with the earlier report [25].

The morphologies of the tantalum films deposited at different substrate temperatures characterized by FESEM are shown in Fig. 2. Fig. 2(a–f) shows surface morphologies of tantalum films at different substrate temperatures. It is clear from FESEM images (Fig. 2(a–f)) that fine grained structure are observed showing smooth characteristics with grain size ranging from 18 nm to 30 nm on the surface. It also can be seen from Fig. 2(a–f) that the grain size along the sample surface has a clear increase from 18 to 30 nm as the substrate temperature is increased from room temperature to 250 °C. Since the grain size is increasing with the increase in substrate temperature it is clear that the FESEM results are in good agreement with the XRD results. Fig. 2(g–l) shows the cross-sectional FESEM images of the tantalum films. It is clear from the figures that the density of the films is high with no voided regions and the thickness of the film increased from 97 nm to 165 nm with the increase in substrate temperature. Our results differ with the work of Dorranian et al. [25] in which a conical structure of tantalum film with larger voided boundaries were observed. The drawback of Dorranian et al. work was large resistance in the films due to voided regions between grains. Charge carries are diffracted at the boundary of grains and cannot continue to drift when there are large voided regions between grains [25]. However, in our work, there are no voided regions in deposited films and the density is high. It should be noted that microstructure plays a critical role in the resulting diffusion barrier performance [29]. Thus, it is expected that the dense nanocrystalline tantalum films prepared by e-beam evaporation technique might possess good performance as a diffusion barrier. Fig. 2(m–r) shows their corresponding EDX spectra. It is clear from Fig. 2(m–r) that the EDX spectra show major peaks for