Electrical, optical, and structural characteristics of Al_2O_3 thin films prepared by pulsed ultrasonic sprayed pyrolysis

S. Carmona-Tellez,[1] J. Guzman-Mendoza,[1] M. Aguilar-Frutis,[1,a)] G. Alarcon-Flores,[1] M. Garcia-Hipolito,[2] M. A. Canseco,[2] and C. Falcony[3]
[1]*CICATA-IPN, Miguel Hidalgo, 11500 Mexico D.F., Mexico*
[2]*Instituto de Investigaciones en Materiales, Universidad Nacional Autónoma de Mexico, Coyoacan, 04510 Mexico D.F., Mexico*
[3]*CINVESTAV-IPN, Apdo. Postal 14-740, 07000 Mexico D.F., Mexico*

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The optical, structural, and electrical characteristics of aluminum oxide thin films deposited by pulsed ultrasonic sprayed pyrolysis are reported. The films are deposited on crystalline silicon at temperatures from 400 to 550 °C using a chemical solution of aluminum acetylacetonate, as source of aluminum, and *N*, *N*-dimethylformamide, as solvent. A H_2O-NH_4OH mist is supplied simultaneously during deposition to improve the films' properties. The results showed that the properties of the as deposited films depended strongly on the number of pulses used and on the substrate temperature. The thickness of the films is under 300 Å and the best films' properties showed an index of refraction close to 1.6 and a root mean square surface roughness of about 7.5 Å in average. Infrared spectroscopy shows that SiO_2 is observed at the interface with silicon of the Al_2O_3 films and seemed to play, as expected, a dramatic role in the electrical characteristics of the interface. Films with a dielectric constant higher than 8 and an interface trap density at midgap in the 10^{10} eV^{-1} cm^{-2} range are obtained. Films deposited with three pulses and at 550 °C are able to stand an electric field up to 4 MV/cm.
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I. INTRODUCTION

Aluminum oxide (Al_2O_3) thin films deposited by physical or chemical methods actually find several applications in different areas of technological research. This is mainly because of their properties such as high mechanical resistance, chemical and thermal stability, and high electrical resistivity. In the microelectronics industry, Al_2O_3 is considered a good candidate to replace silicon oxide (SiO_2) as complementary metal-oxide-semiconductor transistor gate dielectric. Actually, for further advance in silicon microelectronics, alternative thin films are strongly demanded. In this application, the main goal is to get an alternative high-*k* gate dielectric with a low "equivalent oxide thickness" (EOT). In addition, for these alternative oxides, good thermodynamic stability and high interface quality on silicon are necessary.[1,2] Besides Al_2O_3, some of the most studied oxides are HfO_2,[3] La_2O_3,[4] Y_2O_3,[5] ZrO_2,[6] etc. These oxides have dielectric constants higher than 10 (SiO_2 has a dielectric constant of 3.9). Al_2O_3 shows high dielectric strength (a dielectric constant close to 10), good thermodynamic stability up to high temperatures, and high interface quality when deposited on silicon. Furthermore, Al_2O_3 films are amorphous when the deposition temperature is of the order of or lower than 700 °C. All these properties are considered favorable when synthesizing alumina thin films.[2,7–14] Al_2O_3 films with good characteristics have been obtained when sophisticated techniques such as atomic layer deposition[7] (ALD), metal organic chemical vapor deposition,[8] (MOCVD), or pulsed laser deposition[9] (PLD) are used. For example, low leakage currents of about 1 nA/cm^2 at an applied electric field of 2 MV/cm are observed in alumina films with 120 Å thickness using ALD. Under the best experimental conditions, Al_2O_3 films deposited by MOCVD show a leakage current of 10 nA/cm^2 at an equivalent oxide thickness of 3.6 nm. The films of Al_2O_3 deposited by PLD on silicon have shown an interface trap density in the range of 1.2×10^{10} eV^{-1} cm^{-2} when deposited at room temperature. Al_2O_3 films grown by rf magnetron sputtering[10] at room temperature and 200 and 300 °C showed that the best dielectric breakdown strength was of 2.1 MV/cm. In this latter case, the thickness of the films ranged from 45 to 130 nm. However, the spray pyrolysis technique is the only inexpensive technique that has been used to obtain high quality alumina thin films with properties similar or superior to the ones obtained with some of the most elaborated methods.[15–17] In previously reported work, 100 nm thick alumina films deposited with the spray pyrolysis technique were obtained with dielectric strengths of about 5 MV/cm, interface trap densities in the range of 10^{11} eV cm^2, and a dielectric constant in the range of 7.9. It was shown that the addition of a mist of H_2O improved the electrical, structural, and optical properties of the films.[16–18] More recent reports have shown that the addition of a source of nitrogen during deposition of the films can improve their properties.[2,5] In the present work, we used the pulsed sprayed ultrasonic method to obtain alumina thin films as thick as 300 Å. From our best information, no previous attempts had been performed to obtain alumina films with excellent properties in this range of thickness with any low cost deposition technique such as spray pyrolysis. The films were pulsed

a)Electronic mail: maguilarf@ipn.mx.

