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Pulsed laser deposition of nasicon thin films

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Abstract

We have deposited NASICON (*Na Super Ionic CON*ductor) films of thicknesses ranging from 100 to 600 nm, by Pulsed Laser Deposition (PLD), on SiO_2/Si substrates. The deposition rates varied from 6×10^{-3} to 0.08 nm/pulse depending on the conditions employed. XPS measurements show that all elements are transferred from the target to the substrate and that the thin film composition is very close to that of the target. Film morphology depends upon the laser energy density at the target. Films deposited at about 2 J/cm^2 show a rough surface and a columnar structure. At lower energy density (400 mJ/cm^2), a smoother surface is obtained. Electrical measurements show good ionic conductivity, and that the films are suitable for the fabrication of electrochemical gas sensors.

1. Introduction

In the last few years there has been an increasing interest in the development of solid state sensors [1,2]. Among these are electrochemical cells based on advanced ionic conductors, such as NASICON (*Na Super Ionic CON*ductor), for the detection of gases [3]. NASICON is a ceramic formed by a solid solution of $NaZr_2(PO_4)_3$ and $Na_4Zr_2(SiO_4)_3$ of average composition $Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$, where x varies between 0 and 3. Conductivity depends on the value of x, and is maximal for $x \approx 2.2$. Until recently, all electrochemical gas sensors using solid ionic conductors were made either from bulk material or thick films.

The fabrication of solid state sensors by batch processes such as those used in the microelectronics industry is a desirable objective; it requires the ability to deposit thin films of ionic conductors. Because of its complex stoichiometry, it is difficult to deposit NASICON by conventional evaporation or sputtering techniques. Recently, NASICON films in the silica limit ($x = 3$) and containing silicon and phosphorus ($x < 3$) have been deposited by magnetron sputtering [4,5]. For the more general case, including $x \approx 2.2$, the film composition is difficult to control. Loss of Na and P from the target to the substrate is observed when such films are sputtered [6]. As the conductivity of NASICON is very dependent upon the compo-

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