Two step synthesis of $TlBa_2Ca_2Cu_3O_x$ films on Ag substrates by spray pyrolysis of metal-acetylacetonates

L. Pérez-Arrieta and M. Aguilar-Frutis
Centro de Investigación en Ciencia Aplicada y Tecnología Aplicada, Instituto Politecnico Nacional, Miguel Hidalgo 11500, Mexico, D.F. Mexico,
e-mail: mlp.arrieta@yahoo.com.mx, mafrutis@yahoo.es

J.L. Rosas-Mendoza, C. Falcony, and M. Jergel
Physics Department, Centro de Investigación y Estudios Avanzados, Instituto Politecnico Nacional, Apartado Postal 14-740, 07300, Mexico, D.F. Mexico,
e-mail: jrosas@fis.cinvestav.mx; cfalcony@fis.cinvestav.mx; m.jergel@chello.sk

Recibido el 13 de agosto de 2008; aceptado el 24 de noviembre de 2008

A two-step synthesis process was performed to obtain $TlBa_2Ca_2Cu_3O_x$ (Tl-1223) superconductor films. The synthesized films were obtained under different thallium diffusion conditions in a two-zone furnace precursor films were deposited at 550°C on silver substrates using the spray pyrolysis technique from acetylacetonates (2, 4-pentanedionates) as reagents. Second, a thallium diffusion process is carried out to incorporate Tl in the films. For this task Tl_2O_3 pellets, as a Tl source, were used at 750°C. Different oxygen flow rates at atmospheric pressure were used in order to get the thallous oxide p(Tl_2O) partial pressure in the range of 6.9×10^{-4} to 6.1×10^{-2} atm. The Tl-1223 phase was obtained in all cases, though for a low p(Tl_2O), the films presented the $BaCuO_2$ phase mixed with the Tl-1223 one. Critical temperature (T_C) values for these films were in the range of 90 to 102 K.

Keywords: Superconductors; thin films; spray pyrolysis.

El proceso de síntesis de dos pasos fue realizado para obtener películas superconductoras de $TlBa_2Ca_2Cu_3O_x$ (Tl-1223). Las películas sintetizadas fueron obtenidas bajo diferentes condiciones de difusión de talio en un horno de dos zonas. En el primer paso, las películas precursoras se depositaron a 550°C sobre substratos de plata usando la técnica de rocío Pirólitico a partir de acetilacetonatos (2,4-pentanedionatos) como reactivos. En el segundo paso, se llevó a cabo un proceso de difusión de talio para incorporar Tl en las películas. Para ésta tarea, se usaron pastillas de Tl_2O_3 como fuente de talio a 750°C. Se usaron diferentes razones de flujo de oxígeno a presión atmosférica para obtener presiones parciales de óxido taloso p(Tl_2O) en el intervalo de 6.9×10^{-4} to 6.1×10^{-2} atm. La fase Tl-1223 se obtuvo en todos los casos, aunque a bajas presiones parciales de p(Tl_2O), las películas presentan la fase $BaCuO_2$ mezclada con la fase Tl-1223. Los valores de temperatura crítica para estas películas se encuentran en el intervalo de 90 a 102 K.

Descriptores: Superconductores; películas delgadas; rocío pirolítico.

PACS: 74.78.Bz; 74.62.Bf; 74.72.Jt; 81.15.Rs

1. Introduction

Tl-Ba-Ca-Cu-O high-temperature superconducting (Tl-HTS) films have been the subject of extensive research activity. In particular the Tl-1223 phase is of interest because of its electronic properties, high-critical current, low anisotropy, and great flux pinning [1]. Tl-1223 films have been reported to have a zero-resistance temperature up to 112 K and a critical current density, under zero magnetic field, of the order of 10^4 A/cm^2 at 77 K [2]. However, Tl-based superconductors have been reported to have difficulties in controlling the formation of single superconducting phase due to their volatility at the typical growth temperatures used for these cuprates. In order to overcome this difficulty, *ex-situ* growth methods have been used extensively for the synthesis of Tl-HTS thin films such as the two-step process. This process consists in first depositing a non-superconducting precursor film with the required Ba-Ca-Cu oxides composition onto a substrate, by either d.c. or rf sputtering, screen printing, spray pyrolysis or any other deposition technique [3,4,5], and then placing this precursor film in a furnace for thallination and conversion to a Tl-HTS material phase. Chemical processes such as spin coating have also been used to deposit the precursor films, but the preparation time is too long, and requires a post annealing treatment at 600 °C for several hours (10 h) [6]. Similarly, the screen-printing method needs several steps, first the sol and later the gel formation with posterior conversion into oxides at high temperatures (800 °C) for very long times (50 h) [7]. Spray pyrolysis is a very simple, low-cost deposition technique that has been used extensively for the synthesis of Tl-HTS precursor films using metal nitrate solutions, although in this case, several deposition cycles (heating – cooling) are required to obtain the adequate precursor film synthesis and thickness [8,9]. Similar thermal treatments are required in precursor films deposited by electrodeposition techniques.

Thallium diffusion has been performed in either one-or two-zone furnaces in order to obtain thallium based superconductor films. A one-zone furnace has been used to obtain superconductor films consisting of a mixture of Tl-2212 and Tl-2223 phases [10,11]. In these works, precursor films had been previously deposited by spray pyrolysis from acetylace-