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Effect of Au/SiO_2 substrate on the structural and optical properties of gallium nitride grown by CVD

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Abstract. **The improvement of the growth of thick GaN films using a fused silica wafer covered with a thin gold layer by chemical vapour deposition at 800 °C is reported. In order to compare the surface properties, crystalline quality, micromilling performance and luminescence, the characterization of a GaN film grown on a silicon wafer is presented as well. The different morphologies of the surface observed on the GaN films are compared on each substrate and the resulting microstructures are presented in detail. High resolution TEM images of the GaN films show the main crystallographic planes characterizing these structures. The wurtzite structure was determined for each sample using the substrates of Au/SiO_2 and Si (100) from the XRD patterns. Also, the re-deposition effect after ion milling of the GaN films is reported. The performance of ionic beam on the surface of the GaN thick films for the geometries patterning of rectangular, circular and annular with two different ion doses was compared. Cathodoluminescence spectra showed that the top surfaces of the samples emit strong UV emissions peaked at 3·35 and 3·32 eV which are related to the Y_4 and Y_6 transitions.**

Keywords. **Gallium nitride; gold layer; fused silica; silicon; substrate.**

1. Introduction

Gallium nitride (GaN) and their alloys (InGaN–AlGaN) have found important applications due to their broad band gap (3·4 eV) suitable for high power electronics, light emitting diodes, sensors and solar cells (Hwa-Mok *et al* 2004; Dong *et al* 2009; Chen *et al* 2012; Chang-Ju *et al* 2013). Initially, the gallium nitride was developed as powder material but the development of their applications in optoelectronics gradually decreased the attention of researchers.

The technical difficulties inherent to the handling as a powder and the control of their physicochemical properties, allowed the development of thick or thin films of these compounds, which was more practical and reproducible. As a result, GaN has established more remarkable results on films technology and their wide expansion into the optoelectronics devices at blue and ultraviolet wavelengths (Li *et al* 2006; Sun *et al* 2010).

Currently, the high-quality GaN films have been grown through several techniques such as metal–organic chemical vapour deposition (MOCVD) and molecular beam epitaxy (MBE) (Hughes *et al* 1995; Chul-Woo *et al* 1999; Martinez-Criado *et al* 2000; Hersee *et al* 2006; Thillosen *et al* 2006; Richter *et al* 2008; Sobanska *et al* 2012). By means of these growth methods, the improvement of luminescence efficiency in the GaN-based devices was characterized on various substrates. Despite of the high dislocation density obtained, the quality of GaN has been improved using the sapphire substrate. However, the search of a suitable substrate for this nitride is still in discussion and other candidates have been proposed for GaN epitaxy such as ZnO, β-SiC, BP, GaAs, GaP, Si, $MgAl_2O_4$, MgO, γ-$LiAlO_2$, ZrN, ScN and TiN (Liu and Edgar 2002).

Besides of the research on the growth techniques for the gallium nitride, the influence of the substrate on the crystalline, electron, optical and compositional properties of the GaN deposition is still in discussion, which motivates to discover new routes to improve the quality of gallium nitride films. The present work reports the results on the surface morphology, crystalline structure, milling pattern and optical emission of GaN films grown by chemical vapour deposition at 800 °C. To compare the GaN properties under different substrates, a fused silica wafer covered with a thin gold layer (Au/SiO_2) and a wafer of monocrystalline silicon (Si) are used. Using scanning electron microscopy (SEM), the formation of segregates and irregular polyhedral in the surface of GaN films on the substrates of Au/SiO_2 and Si were found. By means of transmission electron microscopy (TEM), high

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$In_xGa_{1-x}N$ fibres grown on Au/SiO_2 by chemical vapour deposition

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Abstract. The growth of $In_xGa_{1-x}N$ films ($x = 0{\cdot}1$ and $x = 0{\cdot}2$) on a thin gold layer (Au/SiO_2) by chemical vapour deposition (CVD) at 650 °C is reported. As a novelty, the use of a Ga–In metallic alloy to improve the indium incorporation in the $In_xGa_{1-x}N$ is proposed. The results of high quality $In_xGa_{1-x}N$ films with a thickness of three micrometres and the formation of microfibres on the surface are presented. A morphological comparison between the $In_xGa_{1-x}N$ and GaN films is shown as a function of the indium incorporation. The highest crystalline $In_xGa_{1-x}N$ films structure was obtained with an indium composition of $x = 0{\cdot}20$. Also, the preferential growth on the (002) plane over $In_{0.2}Ga_{0.8}N$ was observed by means of X-ray diffraction. The thermoluminescence (TL) of the $In_xGa_{1-x}N$ films after beta radiation exposure was measured indicating the presence of charge trapping levels responsible for a broad TL glow curve with a maximum intensity around 150 °C. The TL intensity was found to depend on composition being higher for $x = 0{\cdot}1$ and increases as radiation dose increases.

Keywords. Nitride of group-III; indium–gallium nitride; chemical vapour deposition.

1. Introduction

Nitrides of group-III (GaN–InN–AlN) have gained attention due to their potential in optoelectronic applications such as laser diodes (Ponce and Bour 1997), solid-state lighting (Morgan and Zhizhen 2002), transistors (Deb *et al* 2006), UV sensors (Roberts *et al* 2002) and the new generation of solar cells (Jani *et al* 2007; Lestrade *et al* 2011).

The ternary alloy InGaN is a novel material with an important performance in the new optoelectronic devices such as light emitting diodes (LEDs) and blue laser diodes (LDs). Specifically, the InGaN acts as the active layer (Nakamura 1999) responsible to produce emissions with wavelengths at the UV, blue, green, amber and red. According to Vegard's law, it is possible to change the energy value of the band gap from 3·4 (GaN) to 0·7 eV (InN) by varying the indium composition ($0 < x < 1$) in the $In_xGa_{1-x}N$ compound, which modify the optical properties of this nitride range from UV, VIS to IR regions of the electromagnetic spectrum (McCluskey *et al* 1998; Van de Walle *et al* 1999).

Since most of the studies have been carried out for the binary materials GaN and InN, the knowledge of the InGaN ternary is still scarce and incomplete. Also, the manufacture of InGaN needs to overcome technical difficulties that currently exist, especially, the optimal growth conditions and suitable substrates to form high-quality InGaN solid-solutions (Ambacher 1998).

In the literature, there is a substantial amount of studies on the nitride compounds prepared as films and powders (García *et al* 2002). Since the GaN, InN and InGaN films have found more potential applications, the properties of these materials are widely investigated. Several growth techniques to produce these materials into films have been considered including CVD, MOCVD, MBE and MOVPE methods (Matsuoka *et al* 1992; Dupuis *et al* 1999; Red Kin *et al* 2004; Mueller *et al* 2005; Stoica *et al* 2006).

Parallel to the development of methods to obtain InGaN films, the substrate used is an essential issue, since a lattice mismatch between the substrate and the nitride may have a significant disadvantage in producing semiconductor materials with high quality and reproducibility. Commonly, sapphire (Dong-Joon *et al* 2001; Wu *et al* 2003; Hahn *et al* 2011) has been used due to its high order of symmetry and smaller lattice differences with respect to nitrides. Also, other substrates have been used to grow InGaN films such as 6H-SiC, AlN, 3C-SiC, GaAs, ZnO, $LiGaO_2$ and MgO (Liu and Edgar 2002).

From the above, it follows that the parameters influencing the growth, alloy composition, substrate and the chosen method have a decisive effect on the properties of the InGaN solutions. Thus, the multiple variables involved motivates the research and development of novel synthesis techniques to find the optimal conditions for producing high quality InGaN materials.

In the present work, the growth of $In_xGa_{1-x}N$ films with an indium composition of $x = 0{\cdot}1$ and $x = 0{\cdot}2$ using

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ESTUDIO DEL EFECTO DE DIFERENTES SOPORTES MIXTOS EN LA ACTIVIDAD CATALÍTICA Y LAS CARACTERÍSTICAS ESTRUCTURALES DE CATALIZADORES DE $Bi_2Mo_XW_{1-X}O_6$

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Palabras clave: actividad catalítica, oxidación de monóxido de carbón, óxidos mixtos, catalizadores soportados

RESUMEN

Se preparó una serie de catalizadores de $Bi_2Mo_xW_{1-x}O_6$ soportados en Al_2O_3-SiO_2, SiO_2-TiO_2 y carbón activado (C*). El propósito principal fue comparar el uso de diferentes soportes, así como también valorar el efecto de la temperatura de síntesis de los catalizadores en términos de su eficacia y de conversión en la reacción de oxidación de monóxido de carbono (CO) y en su temperatura de activación. La fase activa de los catalizadores, $Bi_2Mo_xW_{1-x}O_6$, se sintetizó a partir de los compuestos de alta pureza $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, $(NH_4)_6W_{12}O_6{\cdot}H_2O$, $Bi(NO_3)_2{\cdot}5H_2O$, empleando el método de co-precipitación química, y posteriormente soportada por el método de impregnación en Al_2O_3-SiO_2, SiO_2-TiO_2 ó C*. Estos sistemas catalizador/soporte se caracterizaron mediante las técnicas de difracción de rayos X (XRD), microscopía electrónica de barrido (SEM) y área superficial (método BET). Con respecto a la actividad catalítica, el compuesto con mayor eficacia en la conversión de CO a CO_2 fue $Bi_2Mo_xW_{1-x}O_6$ /C*, que se preparó a 500 °C; se activó a 125 °C logrando una conversión de 90 %. Se concluye que existe un efecto de la temperatura de síntesis y del tipo de soporte que influye en los diferentes valores de actividad catalítica encontrados.

Key words: catalytic activity, carbon monoxide oxidation, mixed oxides, supported catalyst

ABSTRACT

A series of $Bi_2Mo_xW_{1-x}O_6$ catalysts supported on Al_2O_3-SiO_2, SiO_2-TiO_2 and activated carbon (C*) were synthesized. The aim was to compare the different supports and calcination temperature of catalysts, studying their efficiency and activation temperature in the CO oxidation reaction. The catalysts active phase, $Bi_2Mo_xW_{1-x}O_6$, was made by means of chemical precipitation procedure starting from high purity $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, $(NH_4)_6W_{12}O_6{\cdot}H_2O$, $Bi(NO_3)_2{\cdot}5H_2O$ compounds, which