Communication

Novel Approach to Synthesize Nanostructured Gallium Oxide for Devices Operating in Harsh Environmental Conditions

Badriyah Alhalaili ¹, Ruxandra Vidu ^{2,3,*} , Ileana Nicoleta Popescu ^{4,5,*} , Dhanu Radha Samyamanthula ⁶ and M. Saif Islam ³

- ¹ Nanotechnology and Advanced Materials Program, Kuwait Institute for Scientific Research, P.O. Box 24885, Safat 13109, Kuwait; bhalaili@kisir.edu.kw
 - ² Faculty of Materials Science and Engineering, University POLITEHNICA of Bucharest, 060042 Bucharest, Romania
 - ³ Department of Electrical and Computer Engineering, University of California Davis, Davis, CA 95616, USA; sislam@ucdavis.edu
 - ⁴ Faculty of Materials Engineering and Mechanics, Valahia University of Targoviste, 13 Aleea Sinaia Street, 130004 Targoviste, Romania
 - ⁵ The Scientific and Technological Multidisciplinary Research Institute (ICSTM-UVT), Valahia University of Targoviste, 13 Aleea Sinaia Street, 130004 Targoviste, Romania
 - ⁶ Water Research Center, Kuwait Institute for Scientific Research, P.O. Box 24885, Safat 13109, Kuwait; vdhanuradha@kisir.edu.kw
- * Correspondence: rvidu@ucdavis.edu (R.V.); pinicoleta24@yahoo.com or ileana.nicoleta.popescu@valahia.ro (I.N.P.)

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Abstract: The importance of Ga₂O₃-based material for harsh environmental applications has attracted the interest of researchers in exploring various fabrication and growth techniques of Ga₂O₃-based nanomaterials using effective and low-cost processes. Herein, a demonstration to improve the wettability of liquid gallium on a rough silicon surface is presented. To control the roughness process, the silicon surface was patterned and groove-shape structures on the silicon were created using a photoelectrochemical (PEC) etching technique. Gallium oxide nanostructures were grown by thermal oxidation from liquid Ga in the presence and the absence of a silver thin film used as a catalyst. Scanning Electron Microscopy (SEM) was used to observe the morphology of the nanostructures grown on the roughed surface of the silicon substrate. The conformal deposition of Ga₂O₃ nanostructures inside the grooves of the PEC etched silicon surface was observed. The presence of Ag catalyst was found to completely change the morphology of Ga₂O₃. This method is recommended for the sustainable and low-cost synthesis of nanostructured gallium oxide for applications, including gas sensing.

Keywords: Ga₂O₃ nanostructure; liquid Ga wettability; thin films; PEC etching; sustainable coating; SEM analysis; gas sensors; harsh environmental applications

1. Introduction

A considerable decrease in natural resources [1,2] has been observed in recent years, which requires immediate attention from researchers and the industry community. Environmental pollution is so severe at the moment that it requires significant technological advances that must be made as soon as possible. In this regard, the design and development of novel engineered materials exploiting micro/nanodevice fabrication and integrating various processes in clean technology [3–7] are the most significant ways to reduce pollution.

Among the various sources of pollution, significant gas pollution is caused both by economic activities, which generate air pollutants resulting from industrial processes (industrial pollutants) [3,4,8,9] and pollution resulting from the emissions of gases/noxious substances from vehicles, aircraft, railways or naval vehicles, or other toxic gases from indoor/outdoor environments [10–12]. Therefore, there is a need to develop sensors

and devices based on materials with excellent properties at high temperatures, which work in harsh environmental conditions [13]. In order to quickly and efficiently detect various gases in aggressive environments (i.e., high temperature, pressure and radiation, corrosive, explosive or toxic environment, etc. [13]), gas sensors made of materials with the required structural and electronic properties are essential for the sustainable detection of gas emissions in these particular harsh conditions [14].

Among the semiconductor materials used in the sensing industry, gallium oxide (Ga_2O_3) offers exceptional structural stability and excellent chemical resistance at very high temperatures (above 1000 °C) [15], thermal stability (most stable at temperatures above 750 °C) and has relatively low thermal conductivities as compared to SiC, sapphire and GaN [16]. β - Ga_2O_3 gas sensors could be used in both oxidizing and reducing atmospheres. It can be used to detect organic vapors at temperatures under 700 °C through surface redox reactions [17]. Fleischer et al. [18] demonstrated that Ga_2O_3 gas sensors have stable electrical responses to 1% H_2 in synthetic air and 5% H_2 in a nitrogen atmosphere at 600 °C. For the detection of reducing gases (e.g., H_2 , ethanol, CO, CH_4 , etc.) various researchers have used β - Ga_2O_3 thin films [16,18,19].

Recently, there has been growing interest in investigating efficient high-k gate dielectrics for flexible and stretchable electronics which are capable of providing a superior gate capacitance that could replace the current gate oxides, such as silicon dioxide. Effective dielectrics require a wide energy band-gap, a large gate leakage current, good interface quality, excellent process compatibility and high stability with the Si surface [20]. Different gate dielectric materials have been used for semiconductor devices, such as silicon dioxide (SiO_2), silicon nitride (Si_3N_4), aluminum oxide (Al_2O_3), zirconium oxide (ZrO_2), hafnium oxide (HfO_2), aluminum nitride (AlN) and gadolinium oxide (Gd_2O_3) [21–25]. The gate dielectric of a metal oxide semiconductor (MOS) capacitor is essential for electrical devices due to its influence on the capacitance of the gate. To obtain a high gate capacitance, a material with a high dielectric constant should be used. Ga_2O_3 is a promising gate dielectric material with a dielectric constant of 9.93 and 10.2 [26], which is much higher than the dielectric constant of silicon dioxide (3.9) [27]. Ga_2O_3 has a high resistivity to carriers that tunnel at the interface between the gate and silicon, owing to its higher dielectric constant [28]. In addition, Ga_2O_3 has a wide bandgap (~4.9 eV) and high chemical and thermal stability, which makes Ga_2O_3 an interesting material in the study of high-k gate dielectrics.

Ga_2O_3 thin film has been demonstrated as a gate dielectric by a number of research groups [29–35]. However, so far, no study has been conducted to enhance the oxidation of liquid gallium's wettability to improve the coating of Ga_2O_3 on the surface of the substrate with inexpensive and sustainable processes. It has been shown by our group that coating the surface with a thin film of metallic material, such as silver, prior to the oxidation process leads to a more homogeneous coating and a denser nucleation of Ga_2O_3 due to the low contact angle [36]. The contact angles of Ga on a silver film and silicon substrate are 30° and 73.9° [37,38], respectively, resulting in a better wetting of Ga on Ag surface and the uniform growth of Ga_2O_3 nanowires. However, this process requires the sputtering technique, which increases the cost of the fabrication. Our approach is to explore other versatile and sustainable techniques to enhance the wetting property of liquid Ga on the surface of different substrates. Thermal oxidation is an inexpensive method to grow Ga_2O_3 thin film or nanowires, but the poor wettability of Ga on Si substrates limits the use of this process for electronic applications. To study the wettability of Ga and the growth morphology of Ga_2O_3 on a Si surface with controlled roughness, the first step was to produce groove-like structures on a silicon surface using a photoelectrochemical (PEC) etching technique to fabricate structures with a definite pattern on the surface of the silicon substrate. Then, the wettability properties of Ga were studied on this patterned Si surface in the presence and the absence of a Ag catalyst.

2. Materials and Methods

The substrate was n-type (100) P-doped Si wafer with a thickness of 500 μm and a resistivity of 3 to 5 $\Omega\cdot\text{cm}$. The Si wafer was cleaned using acetone and methanol for 5 min each in an ultrasonic bath. Photoelectrochemical etching was used to fabricate grooves on silicon surface according to the process developed in previous work [39]. The silicon surface was coated via low pressure chemical vapor deposition (LPCVD) with Si_3N_4 , which was used as a hard photomask. The substrate was patterned and etched by KOH to introduce v-groove in the silicon surface for deep etching by PEC etching technique.

Then, the substrate was subjected to the PEC etching process. A 2A high-power multimode fiber-coupled laser with a wavelength of 975 nm (Qphotonics LLC, Ann Arbor, MI, USA) was used to illuminate the wafer from the backside while etching occurs on the front side of the wafer. An electrochemical technique in HF-based electrolytes was used to fabricate deep microstructures in the patterned substrate. The applied voltage was controlled by CHI 660 electrochemical workstation (CHI Instruments Inc., Austin, TX, USA). The Teflon electrochemical cell contained the electrolyte consisting of $\text{C}_2\text{H}_5\text{OH}$ 99.9% and DI H_2O with 5% HF in a volume ratio of 1:1. Finally, after PEC etching process, the sample was rinsed in deionized water and then dried with nitrogen. Another set of substrates were coated with a layer of 5 nm Ag as a catalyst by Lesker sputtering.

To grow Ga_2O_3 nanostructures, 0.2 g of gallium [(Ga) (purity 99.999%)] was dripped onto the cleaned PEC-etched silicon surface and on the Ag-coated silicon surface. Figure 1 shows the substrate processing and Ga oxidation process.

Figure 1. Schematic representation of fabrication and oxidation processes of the silicon substrate and gallium, respectively, including the PEC etching process. Both patterned silicon surface, i.e., with and without a Ag catalyst, were coated with Ga and oxidized at 1000 $^{\circ}\text{C}$ in N_2 gas for 1 h.

The samples were exposed to the heat plate to obtain uniform coating of Ga. The substrate was heated at 1000 $^{\circ}\text{C}$ for 1 h in nitrogen flow. All the characterizations were performed at room temperature. Sample was cleaved and analyzed using cross-sectional Scanning Electron Microscopy equipped with a FEI Nova 410 NanoSEM (FEI) system.

3. Results and Discussion

Figure 2 shows the SEM images of gallium oxide on silicon together with two cross-sectional views of the spots marked in the top view image. The Ga–Si phase diagram shows an eutectic at 99.994 at.% Ga and 29.7 °C [40]. Heating the Ga–Si system at 1000 °C and then cooling to room temperature results in droplets formed through melting and dewetting at the eutectic temperature. Due to the dewetting effect of liquid Ga on the Si surface, the coverage of the Ga₂O₃ nanostructures was not homogenous, as shown in Figure 2. Some Ga drops tend to form small droplets in the range of 1 µm on the surface of silicon and not wet it. Ga has high surface tension and does not wet hydrophilic-like surfaces such as silicon. The SEM observation of Ga nanospheres presented in Figure 2b,c suggests a large contact angle (α) close to 180°, which corresponds to low wettability. Under the presence of a small amount of oxygen, some of these Ga nanospheres are oxidized and transformed into nanowires (Figure 2c); however, some of them remained as perfect liquid Ga spheres in the range of 1 to 2 µm (Figure 2b).

Figure 2. SEM images of Ga₂O₃ nanostructures growth on Si surface; (a) Liquid Ga oxidation on silicon surface leads to nonuniform coating; (b) non-oxidized Ga spheres on Si surface; (c) oxidized Ga nanospheres and formation of Ga₂O₃ nanowires on the surface.

To explore the wetting property of liquid Ga on the surface of silicon, grooves with micro- and nanosize structures were introduced on the silicon surface by the PEC etching technique. Figure 3 shows the SEM images of the top and cross-section views of the PEC etched Si surface, where the diameter and the width of the uniform, concave-shape grooves were $\sim 5.83 \mu\text{m}$ and $\sim 12.10 \mu\text{m}$, respectively.

Figure 3. SEM images of groove-shape structures on silicon surface (top view, (left)) and side view (right) of the Si substrate after PEC etching.

Figure 4 shows the SEM images of the oxidized Ga on the PEC-etched surface of Si in the absence and in the presence of a Ag catalyst.

Figure 4. SEM images of the oxidized Ga on PEC-etched surface of Si in the absence (a,b) and in the presence (c,d) of a Ag catalyst (left: top views; right: cross-section views).

Although Figure 4a suggests that gallium melts and forms melted-like patches on the PEC-etched Si surface, Figure 4b shows that, actually, the liquid gallium does not enter the grooves in the absence of a Ag catalyst. In addition, the liquid gallium oxidation does not take place completely, and the modified surface texture of the silicon actually supports the liquid Ga and does not allow it to enter the grooves in such condition as shown in Figure 4a,b. By comparison, a major change in the growth morphology of Ga_2O_3 nanostructures was observed in the presence of a Ag catalyst, where an increase in the wetting property of liquid Ga was observed. Due to the catalytic action of the Ag layer, Ga_2O_3 was grown inside the grooves of the PEC-etched silicon surface (Figure 4c,d), where there was a conformal Ag layer. In the presence of a Ag catalyst, a dense growth of nanowires was observed on the top surface and inside the grooves (Figure 4d).

The Growth Mechanism of Ga_2O_3 Nanostructures

During the oxidation process, three major steps are involved in the vapour–liquid–solid (VLS) growth mechanism and in the formation of Ga_2O_3 . The growth mechanism is summarized in Figure 5.

Figure 5. Growth mechanism and formation of Ga_2O_3 .

In the first step, liquid gallium oxidizes at the surface of silicon substrate to form gallium (III) oxide (Ga_2O_3) (Equation (1) in Figure 5) [41,42]. In the second step, the Ga_2O_3 solid phase interacts with the gallium liquid phase to form the gallium suboxide (Ga_2O) gas phase (Equation (2) in Figure 5) [43]. In the third step, the Ga_2O vapor phase breaks down into liquid Ga and Ga_2O_3 in the presence of oxygen (Equation (3) in Figure 5) [44,45].

In the presence of a Ag catalyst, the rate of formation of the solid phase of gallium oxide (III), Ga_2O_3 , increases significantly. The dissolution and diffusion of oxygen into solid silver [46] is much higher than in liquid Ga [42]. In addition, it has been shown that the oxygen solubility is very high with increasing temperature [47]. At high temperatures, O_2 dissociation acts as a pool of O atoms supply for Ga oxidation. The O_2 concentration gradient would increase to establish a dynamic equilibrium of Ga–Ag–O at 1000 °C. It could be expected that there would be a high concentration of oxygen atoms near the exposed surfaces of the AgO_x nanoparticles (NPs) and a low oxygen concentration near the Ag/ Ga_2O_3 interfaces. Therefore, more oxygen is dissolved in the silver nanoparticle leading to a continuous supply of Ga_2O (g) and O_2 (g), which increases the formation of the Ag–Ga–O liquid mixture and the growth of Ga_2O_3 (s) to spontaneously form denser nanowires [36,48]. It is important to note that high nitrogen (N_2) pressure (1–2 GPa) and high temperature (1400–1500 °C) is necessary to drive the reaction between Ga and N_2 to form GaN [49], which was not achieved in our experiments. In addition, our material characterization using XRD and XPS did not show the presence of elemental N or GaN [50].

4. Conclusions

The wettability of liquid gallium and its oxidation on a modified surface of silicon was observed in the presence and absence of a Ag catalyst. A photoelectrochemical etching technique was used to modify the silicon surface by creating concave structures on the surface. In the presence of Ag catalyst, it was observed that (i) Ga₂O₃ tends to form a homogenous coating even within the deep, groove-like structure on the Si surface, and (ii) an enhanced growth and a high density of nanowires was formed. Hence, this paper presents the first successful attempt to increase the wettability of liquid gallium on the silicon substrate with controlled roughness using the thermal oxidation process to study the effect of surface properties of the growth substrate on the Ga₂O₃ morphology.

These observations have a significant impact on the manufacturing of electronic devices. Diverse optoelectronics and electronic applications can use this growth process to create homogenous coatings of the thin film under low temperatures or create conformal coatings of nanowires at high temperatures. Further work on assessing the electrical properties of the Ga₂O₃ nanowires grown by this process is ongoing in our group. Such investigations could be further exploited to develop optoelectronic or electronic devices for applications such as gas sensing.

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