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Gallium Arsenide (GaAs) Nanofibers by Electrospinning Technique as Future Energy Server Materials

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Abstract: Gallium arsenide (GaAs) does have superior electronic properties compared with silicon. For instant, it has a higher saturated electron velocity and higher electron mobility. Weak mechanical properties and high production cost are the main drawbacks of this interesting semiconductor. In this study, we are introducing production of GaAs nanofibers by electrospinning methodology as a very low cost and yielding distinct product technique. In general, nano-fibrous shape is strongly improving the physical properties due to the high surface area to volume ratio of this nanostructure. The mechanical and environmental properties of the GaAs compound have been modified since GaAs nanofibers have been produced as a core inside a poly(vinyl alcohol) (PVA) shell. GaAs/PVA nanofibers were prepared by electrospinning of gallium nitrate/PVA solution in presence of arsenic vapor. The whole process was carried out in a closed hood equipped with nitrogen environment. FT-IR, XPS, TGA and UV-Vis spectroscopy analyses were utilized to confirm formation of GaAs compound. Transmission electron microscope (TEM) analysis has revealed that the synthesized GaAs compound is crystalline and does have nano-fibrous shape as a core inside PVA nanofibers. To precisely recommend the prepared GaAs nanofiber mats to be utilized in different applications, we have measured the electric conductivity and the band gap energies of the prepared nanofiber mats. Overall, the obtained results affirmed that the proposed strategy successfully remedied the drawbacks of the reported GaAs structures and did not affect the main physical properties of this important semiconductor.

Keywords: Optical materials, Gallium arsenide, Nanostructured materials, Sol-gel processes, Semiconductors

Introduction

Recently, gallium arsenide (GaAs) semiconductors had come forward to be used as a material of choice by growing sequence of thin or single crystal layers on single crystal substrate which results in high electrical outputs. However, to use GaAs as an efficient energy devices conserving material, there are mainly two manufacturing methods; molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD) which give GaAs in the form of thin films [1-3]. To exploit the premium advantages of the nanostructures, other researchers have been reported production of GaAs nano-sized quantum dots [4,5]. There are some important constrains against utilizing the reported GaAs structures in wide scale; mainly, the mechanical properties, production cost, and the sensitivity of GaAs to the environmental parameters. For instance, it was seen that GaAs wafers are very fragile and rate of breakage is high [6].

One-dimensional (1-D) nanostructure including nanofibers, nanorods and nanotubes have received increasing interest for their superior optical, electrical, catalytic and magnetic properties, based on their low dimensionality and quantum confinement effect, which can be widely exploited as fundamental building blocks for nanoscience and nanodevices [7]. In general, nanofibers do have the highest surface area/volume ratio in the class of 1-D nanostructures, so, nanofibers are the best candidate for nano devices and nano membranes. Consequently, nanofibers of different semiconductor materials showed high energy retentions, because of their size-dependence [8]. Accordingly, we think that GaAs nanofibers will possess prominent physical characters. According to our best knowledge, GaAs semiconductor has not been reported yet in any 1-D shape.

Electrospinning technique has been paid to considerable attention due to production fibers having diameter in the range of few microns to nanometer level and for economical aspect [9,10]. In a typical electrospinning process, electrostatically driven polymer jet is ejected from polymer solution which undergoes bending instability wherein the solvent evaporates and ultra fine stretched fiber are deposited on the grounded collector [11].

In this communication, the main aim was producing GaAs in nano-fibrous form and simultaneously overcoming the

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aforementioned dilemmas of the reported GaAs nanostructures. Therefore, GaAs nanofibers enveloped in PVA polymer were produced. In the proposed core/shell structure, PVA was invoked to modify the mechanical properties of GaAs as well acts as a transparent insulation to protect the semiconductor from the environmental stresses. It is noteworthy mentioning that PVA shield did not affect the optical properties of GaAs compound, the prepared nanofibers have absorbed light, the onset of the absorbance curve is located at 232 nm and it is blue shifted from the bulk value [12], and all the other reported structures [13] due to the novelty of the produced shape. The electric conductivity and band gap energy of the prepared mats have been studied, the obtained results were satisfactory since the prepared nanofiber mats do have good electrical conductivity and band gap energies almost same as GaAs bulk material.

Experimental

Materials

Poly(vinyl alcohol) (PVA) (M_w=65000), gallium (III) nitrate 8-hydrate (99 % purity), arsenic acid (60 wt%), and zinc nitrate were obtained from different chemicals manufacturing company; they were obtained from Dong Yang Chem., Co. (Korea), Showa Chem., Co. (Japan), Wako Chem., Co. (Japan), and Sigma Aldrich (USA), respectively. These chemicals have been used without any further treatment. Triple distill water was used as solvent.

Characterization

Surface morphologies of obtained nanofibers had been studied by Scanning electron microscope JEOL JSM-5900. Samples were osmium coated and analysed for morphological states. To identify the vibrations in functional groups of GaAs enveloped nanofiber; FT-IR analysis was done using a Varian FTS 1000 spectrometer scimitar series (Varian Inc, Co., Australia). The samples were made by pelletising the previously vacuum dried nanofibers with potassium bromide. Surface states of GaAs were analysed by X-ray photon spectroscopy (XPS, AXIS-NOVA, Kratos Inc.) with K as a source of X-rays. The thermal stability of the samples was carried out with a (Pyris TGA Perkin Elmer Inc., USA) by heating in oxygen atmosphere from 30 to 800 °C with a heating rate of 10 °C/min. This analysis was conducted under a continuous oxygen purge of 20 m*l*/min. Transmission electron microscopy was analyzed by JEOL JEM 2010 (TEM) operating at 200 kV, JEOL Ltd., Japan. Small amount of sample was sonicated for 5 min followed by dip coating of 500 mesh copper grid, further on dried to remove excess of non-solvent and observed for analyses. Optical study was investigated by HP 8453 UV-visible spectroscopy system, Germany. The spectra obtained were analyzed by HP ChemiStation software 5890 series. Current-voltage (I-V) measurements were performed by the semiconductor parameter analyzer at room temperature using Cascade Microtech probe station, USA.

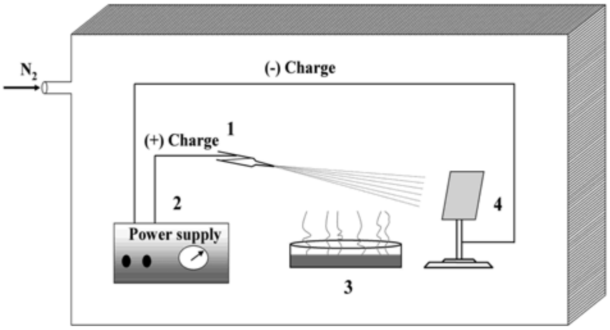
Fabrication of Nanofibers by Electrospinning

A high voltage power supply (CPS-60 K02V1, Chungpa EMT Co., Republic of Korea), was used as a source of electric field. Polymer solution to be electrospun was supplied through a plastic syringe attached to a capillary tip. The copper wire originating from positive electrode (anode) connected with copper pin was inserted into the polymer solution and a negative electrode (cathode) was attached to a metallic collector. Actually, the basic idea of the current study is electrospinning of gallium nitrate/PVA sol-gel in presence of arsenic gas, so, this gas can interact with the gallium nitrate in the flying nanofibers to produce gallium arsenide. The utilized sol-gel has been prepared by mixing 1.2 g gallium nitrate aqueous solution (20 wt/wt%) with 5 g PVA aqueous solution (10 wt%), the solution was electrospun at 15 kV positive voltages, 15 cm distance between the syringe tip and the collector. In order to have full utilization of arsenic gas and to have maximum interaction of arsenic with flying nanofibers and the nanofibers rested on collector we have kept the nanofibers inside the glove box for 12 h as resident time. Moreover, to generate the nanofibers in a safe way as the arsenic is very toxic gas, we devised our experimental procedure in closed glove box under inert nitrogen environment as shown in Scheme 1. The box should be free of any oxygen to avoid oxidation of arsenic gas. The arsenic gas has been synthesized inside the glove box according to the following reactions.

$$H_3AsO_4 + 3NaOH \longrightarrow Na_3AsO_4 + 3H_2O \tag{1}$$

$$2Na_3AsO_4 + 3H_2SO_4 \xrightarrow{ZnNO_3} 3Na_2(SO_4) + 3H_2O + {}^{5}/_{2}O_2 + 2As \tag{2}$$

Typically, 1 m*l* of arsenic acid (H_3AsO_4) has been placed in a petriplate (item 3, Scheme 1), then 17.5 g of zinc nitrate aqueous solution ($ZnNO_3$, 2 N) and 6.63 g sodium hydroxide



Scheme 1. Electrospinning setup used; (1) syringe containing GaNO3/PVA sol-gel, (2) high voltage power supply, (3) petridish containing chemicals to produce arsenic vapor, and (4) grounded collecter to collect the final nanofibers.

aqueous solution (NaOH, 8.16 N) were added to the acid. Finally, few drops of diluted sulfuric acid (H_2SO_4) were added to the reaction media.

The collected nanofibers on collector were vacuously dried for 12 h at room temperature to remove the residual solvents. Other nanofiber mats have been prepared in absence of arsenic gas to properly investigate the effect of arsenic gas.

Results and Discussion

Electrospinning technique involves the use of a high voltage to charge the surface of a polymer solution droplet and thus to induce the ejection of a liquid jet through a spinneret. Due to bending instability, the jet is subsequently stretched by many times to form continuous, ultra thin fibers. PVA does have a wide utilization in producing of ceramic nanofibers via electrospinning methodology for its novel chemical and mechanical characteristics. The hydroxyl groups deployed on the PVA chains have the capability to generate hydrogen bond with many anions (especially those having oxygen atom) which enhances the solubility of the metal salt in PVA solution [14]. At this stage Figure 1(A) shows morphologies of gallium nitrate/PVA electrospun nanofiber mats in absence of arsenic gas. However, Figure 1(B) shows the nanofiber mats which were electrspoun in presence of arsenic gas. As shown in these figures, there is no significant difference between the two mats. However, there is a slight increase in the diameters of the nanofibers obtained in presence of arsenic gas as can be observed from Figure 1(A) and (B). Overall, the obtained average diameter of the peristine nanofibers was in the range of 50 to 900 nm while as its counterpats had an average diameter of 50 to 2000 nm. This increase in the diameter is an indicative of reaction to be occured. The morphologies of the peristine and modified one draws our attention to an intresting findings that in lateral we can see the swelled nanofibers compaied to peristine. This swelled morophology can be atributted to the binding of arsenic vapor on the surfaces of nanofibers durring the electrospinning process which may lead to swell the indivisual nanofibers as compaired with peristine. Meanwhile, it was observed that the color of obtained nanofiber mats in case of arsenic gas treatment were pale brown, while the untreated one was white, this browning in colour can be explained as a formation of GaAs compounds.

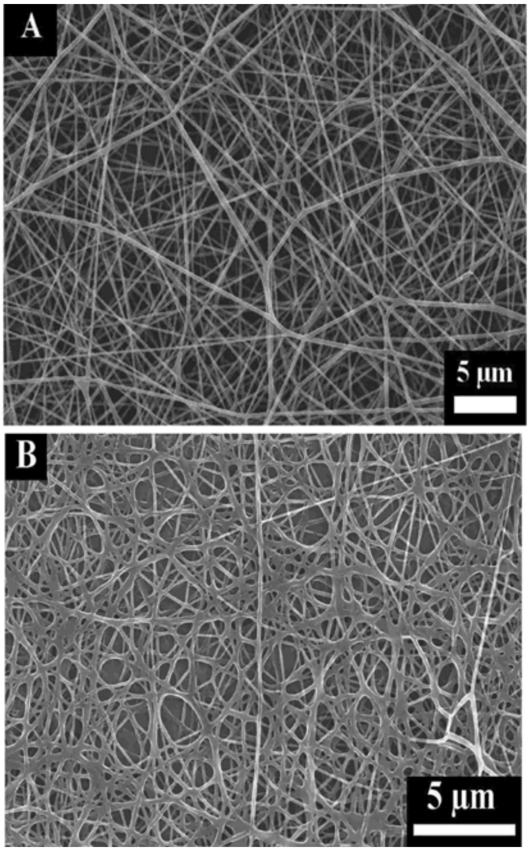


Figure 1. Scanning electron microscopy images electrospun $GaNO_3$/PVA nanofiber mats; (A) in absence arsenic vapor and (B) in presence of arsenic vapor.

Figure 2 represents the obtained FT-IR spectra of gallium nitrate/PVA elsctropun in presence and absence of arsenic gas. As shown in this figure, in a case of the nanofiber mats electrospun in absence of arsenic acid, various stretching bands of nitrate group at 813 and 1033 cm^{-1}, and most intense stretching band of nitrate at 1388 cm^{-1} indicating apparence of nitrate group in the prepared nanofibers [15]. While as the nanofibers which were electrospun in presence of arsenic vapors, the most of the aforementioned peaks indicating *existence* of nitrate group were missed. However, some of the peaks with small intensities were present espacially, at wavelenght of 1388 cm^{-1} can be seen. This presence of small intensity peaks can be atributed due to unreacted nitrate groups in the final product. Overall, results

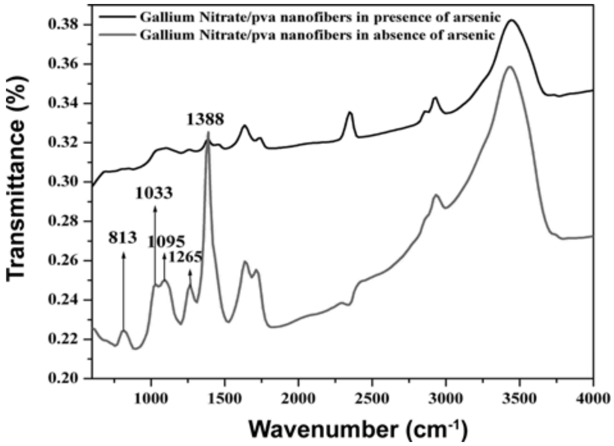


Figure 2. FT-IR spectra of the electrospun $GaNO_3$/PVA nanofiber mats in presence and absence of arsenic vapor.

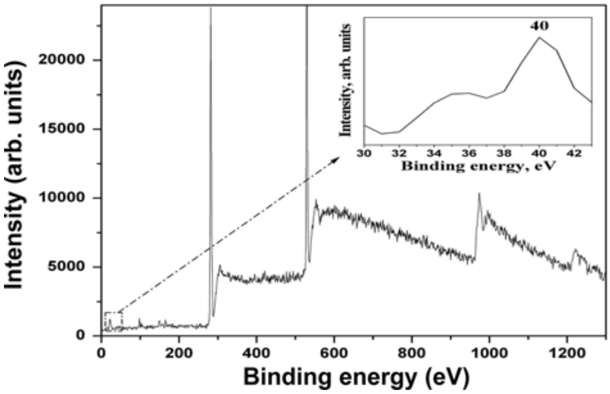


Figure 3. XPS spectra for pure electrospun $GaNO_3$/PVA nanofiber mats with an inset showing higher magnification of a specific area.

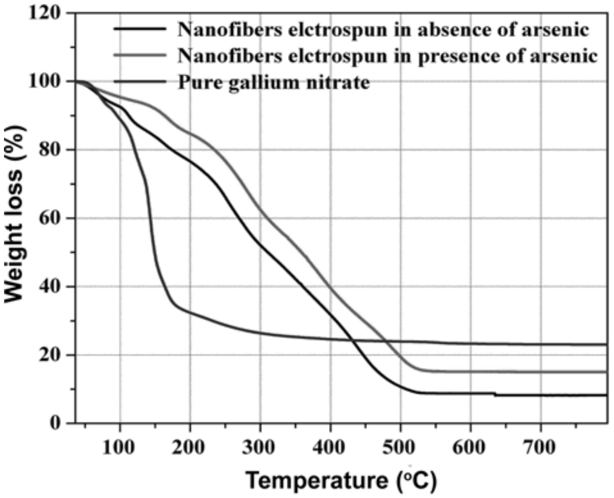


Figure 4. Thermal gravimetric analysis of electrospun $GaNO_3$/PVA nanofiber mats in presence and absence of arsenic vapor and pure gallium nitrate compound.

reveals distinct changes in the chemical composition of the nanofibers have been carried out due to exposure of the nanofibers to the arsenic gas atmosphere.

Pervious studies on GaAs have been revealed that XPS analalysis can detect Ga-As bond as a peak at ~41 eV [16]. This peak revealed to contain As3d5/2 binding energy which bond between gallium and arsenic [17]. Figure 3 represents the XPS spectra for the electropsun nanofibers in presence of arsenic gas. As shown in the inset in this figure, the peak representing As3d5/2 binding energy clearly appears which indicates formation of GaAs. In the main spectra this peak is quite small, with taken into consideration that XPS is a surface analysis technique; one can say GaAs is trivial on the surface. As the FT IR confirmed absence of nitrate group from these nanofibers, so, we can say that the arsenic gas has been reacted with gallium nitrate to for gallium arsenide according to this reaction:

$$GaNO_3 + As \xrightarrow{\quad} GaAs + NO + O_2 \tag{3}$$

Thermal gravimetric analysis has been utilized also to confirm occurance of reaction (3). Figure 4 represents the thermal gravimetric analysis data (in oxyegn atmosphere) for $GaNO_3$/PVA nanofiber mats electrospun in presence and absence of arsenic gas, and pure gallium nitrate compound. As shown in this figure, there are two obvious notes that could be observed. First, the amount of residual inorganic materials is higher than in case of the nanofibers electropsun in presence of arsenic gas, we think this increase of inorganic residuals due to embedding of arsenic metal in the nanofibers which can only be present in the form of gallium arsenide. For instance, the remaining materials in case of the nanofibers electrospun in absence of arsenic vapor is definitly gallium oxide due to complete thermal elemination of PVA since the analysis was conducted in oxygen atmosphere, and also decomopsition of gallium nitrate into oxides form. Formation of gallium oxide can be proofed from the obtained TGA results of pure gallium nitrate compound; as shown in Figure 4, the resiudals in such a case represent 24 % from the total weight, according to the molecular weights and purity of the utilized gallium nitrate, these reiuduals can be only explained as gallium oxide.

Moreover, difference in degradation rates of the two nanofiber mats can also be observed, the nanofibers electropsun in absence of arsenic gas show fast degradation pattern as compared with its counterpart, this change in rates reveals distinct variation in chemical composition between the two nanofibers formulations which confirms taking place of reaction (3).

It is well know that transmission electron microscope analysis (TEM) can be utilized to investigate the crystallinity of the materials [18]. In other words, TEM analysis can easily distinguish between the amorphous and crystalline materials by analyzing the high resolution TEM (HR TEM) and the selected area diffraction pattern (SAED) images. Almost, most of the polymers are amorphous or bad crystalline materials; in this regard one can say that the prepared gallium nitrate/PVA nanofibers can not reveal good crystallinity since gallium nitrate was utilized as a solution. Figure 5 shows the TEM results for the electrospun nanofibers in presence of arsenic gas. As shown in Figure 5(A) which demonstrates HR TEM image; strips of crystalline material clearly appear inside the amorphous PVA polymer. Moreover, SAED image (Figure 5(B)) reveals good crystallinity of these strips since there are no dislocations or imperfections observed in the lattice planes. These results also provide more confirmation that presence of arsenic gas during performing the electrospinning process leads to synthesize GaAs. The main planes in GaAs crystal lattice (i.e. (111), (220) and (311); JCPDS card no. 32-0389) could be detected in the SAED image (Figure 5(B)). Also, we have measured the planar spacing in the HR TEM image (Figure 5(A)), the

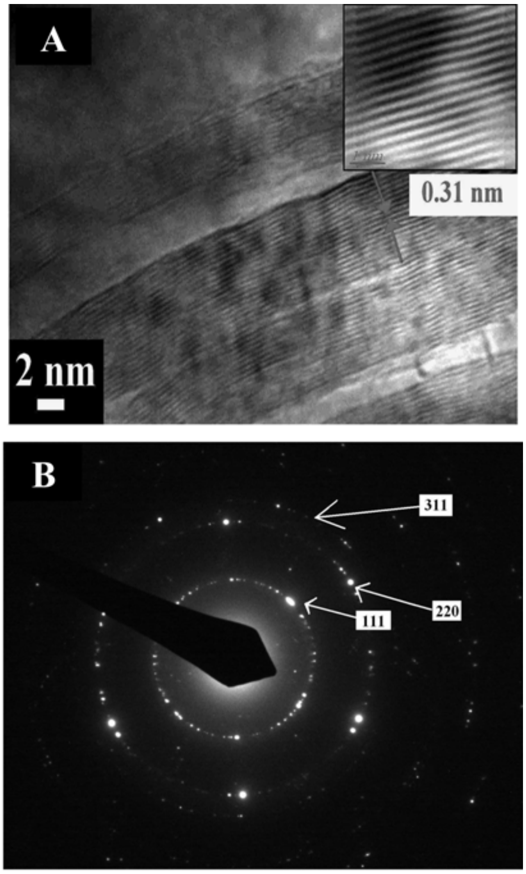


Figure 5. (A) High resolution transmission electron microscopic images of PVA enveloped GaAs nanofiber and (B) selected area electron diffraction pattern of PVA GaAs enveloped nanofiber.

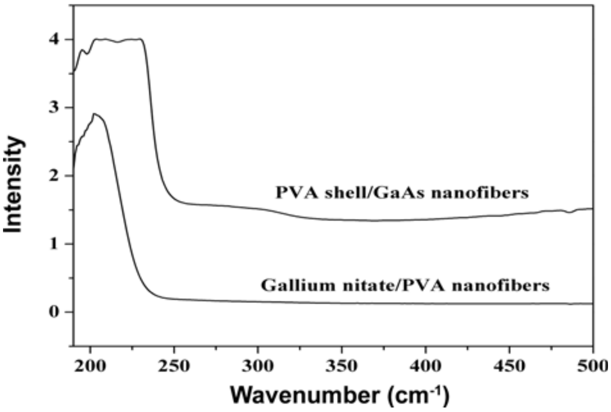


Figure 6. UV-visible spectra of colloidal solution of pure electrospun $GaNO_3$/PVA nanofiber and PVA enveloped GaAs nanofiber.

obtained value almost matches the standard value of GaAs compound. It is noteworthy mentioning that, HR TEM images of the nanofibers electrospun in absence on arsenic gas did not reveal any data, so, we did not offer them. TEM analysis draws our attention to an interesting observation that GaAs semiconductor was synthesized in nano fibrous form, this phenomenon can be explained as arsenic gas interacts with the flying nanofibers so, the produced GaAs does have the same shape. Also, we can say that the gallium nitrate was located in the core of the nanofiber. Therefore, the produced GaAs nanofibers were placed inside the PVA mother fiber. Therefore, one can say that the proposed strategy does have the ability to produce GaAs nanofibers enveloped in PVA shell.

UV-visible is a versatile method to find out the nature of suspended colloid in aqueous solutions. As PVA enveloped GaAs nanofibers, it was assumed that the PVA enveloped GaAs nanofibers could well be dispersed in water after a brief sonication. To begin with analysis, a fixed amount of gallium nitrate/PVA and PVA enveloped GaAs nanofibers were dispersed in ependroff tube and kept under ultrasonic irradiation for 30 min. Figure 6 shows the UV-vis. absorption spectrum of the PVA/gallium nitrate and PVA enveloped GaAs nanofibers. In this Figure one can clearly differentiate between gallium nitrate/PVA and PVA enveloped GaAs nanofibers. In case of optical absorption spectrum of PVA enveloped GaAs nanofibers, it was seen that a broad peak in the range of 200 to 230 nm and the onset of the absorbance curve is located near the start at of 248 nm, is an indicative of blue shift from the bulk value [12]. We think this difference result obtained due to the fibrous shape. Meanwhile, spectrum of the PVA/gallium nitrate was sharp which is completely different from modified one. Overall results indicate the formation of PVA protected GaAs nanofibers.

Actually, GaAs do have superior electronic properties compared with silicon. For instant, it has a higher saturated electron velocity and higher electron mobility, allowing transistors made from it to function at frequencies in excess of 250 GHz [19]. Also, GaAs devices generate less noise than silicon devices when operated at high frequencies. They can also be operated at higher power levels than the equivalent silicon device because they have higher breakdown voltages. These properties recommend GaAs circuitry in mobile phones, satellite communications, microwave point-to-point links, and some radar systems. It is used in the manufacture of Gunn diodes for generation of microwaves. Gallium arsenide solar cells are among the most promising developing technologies available to the spacecraft designer. They offer higher efficiency and increased radiation resistance than silicon (Si) cells. However, there some significant drawbacks of these solar cells, they are heavier, more brittle, and more costly than Si.

The prepared nanofibers can be utilized to overcome some of these drawbacks. For instance, the conventional GaAs solar cells are heavier because of GaAs is more dense than Si, also, to surmount the brittleness problem, the manufactures tend to make little thick layer which possessing better mechanical properties. On the other hand; this is the main

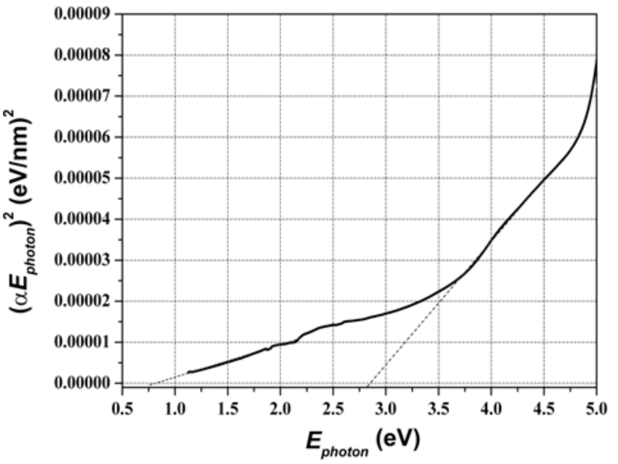


Figure 7. The relationship between the applied voltage and the electric current passed in the nanofiber mats obtained in absence and presence of arsenic.

reason of increasing the cost of the GaAs solar cells. In a case of the prepared nanofibers, the mechanical properties are quit well and the amount of the embedded GaAs is small. Moreover, the manufacturing cost is so cheap compared with the two common methods aforementioned in the introduction section. However, to properly recommend utilizing of the prepared nanofibers not only in solar cells but also as a alternative candidate for GaAs normal structure, two important parameters have to be investigated. These parameters are the electric conductivity of the produced nanofibers and the band gap energy. In other words, we have to confirm that the invoked PVA did not affect the electric conductivity of GaAs, and study the influence of the nano fibrous shape on the band gaps especially previous works have indicated that the optical properties of nanostructures mainly depend on the shape and size [10-21].

Figure 7 shows the IV plot of the pristine $GaNO_3$/PVA and the obtained nanofiber mats in presence of arsenic vapor. As shown in this figure, GaAs shelled nanofibers do have Ohmic behavior since the relationship between the applied voltage and the current is almost linear passing with the origin. This indicates good electric conductivity of these nanofibers. However, in case of $GaNO_3$/PVA nanofibers, one can say that these nanofibers are either nonconductive or badly conductive as shown in Figure 7.

The optical absorbance properties (i.e. the band gap energies) can be studied from UV-visible absorption spectra. For a semiconductor, the absorbance in the vicinity of the onset due to the electronic transition is given by the following equation [22,23].

$$\alpha = \frac{K(hv - E_g)^n}{hv} \tag{4}$$

Where α is the absorption coefficient, hv is the photon energy; E_{Photon}, K is a constant, E_g is the band gap, and n is a value that depends on the nature of the transition (1/2 for a direct allowed transition or 2 for an indirect allowed transition). In this case, n is equal to 1/2 for this direct allowed transition. The band gap can be estimated from a plot of (á E_{Photon})^2 versus photon energy. The linear regions in this plot can be invoked to estimate the band gap energies by calculating the intersection points between these regions and the abscissa (x-axis).

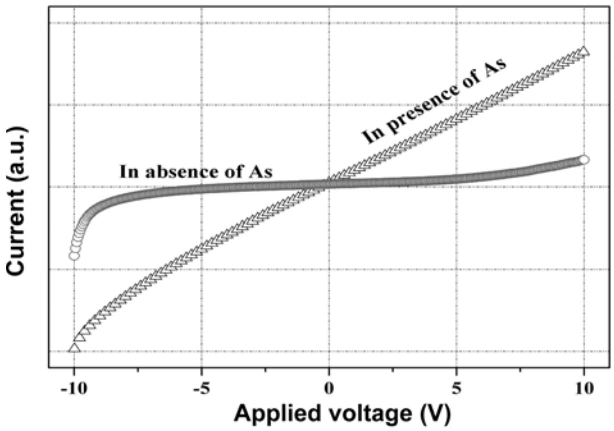


Figure 8. A plot between the photon energy; E_{Photon} versus $(\alpha E_{Photon})^2$ to estimate the band gap energies of the GaAs shelled nanofibers.

Figure 8 shows the relationship between $(\alpha E_{Photon})^2$ and E_{Photon} for the synthesized nanofibers. Extrapolation of the linear regions in the obtained graph can give two E_g values obtained at 0.73 and 2.82 eV. The optical band gap energy difference is 1.59 eV. These band gaps can be explained as the valence and conduction bands. The band gap difference is little higher than the reported GaAs thin film value; 1.43 eV. This indicates that the nano fibrous shape do have trivial effect on the band gaps. According to the obtained results in Figure 7 and Figure 8, we can confidently recommend the prepared GaAs nanofibers to be utilized as an effective alternate for GaAs normal structures.

Conclusion

Electrospinning of a sol-gel consisting of gallium nitrate and PVA in presence of arsenic gas enhances a chemical reaction between arsenic gas and gallium nitrate present in the flying nanofibers. Gallium arsenide semiconductor is the main product of this reaction. The obtained gallium arsenide does have nano-fibrous shape due to formulation of one reactant (gallium nitrate) in this form. Moreover, the formed gallium arsenide nanofibers were placed inside the mother nanofibers as cores surrounded by PVA shell. The proposed strategy does have the ability of producing GaAs semiconductor in a novel shape and also in a form does have better mechanical properties compared with the reported GaAs nanostructures. Also, protection of the produced GaAs nanofibers via PVA shell will strongly recommend this novel shape to be widely utilized in various applications due to overcoming the environmental stresses problems. As the conventional GaAs structures doing have good electric conductivity and proper band gap energy, these two important factors should be satisfied to recommend any proposed structure of GaAs material to be used in GaAs applications. The obtained results in this study indicated good electric conductivity and band gap energy almost same as the reported band gap energy of the bulk GaAs materials. Accordingly, we think the prepared GaAs can be safely exploited in the solar cells and other fields depending on solar energy.

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