Highly Easy and Low Cost Fabrication of Graphite-based Flexible Transparent Conducting Film

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Abstract. We demonstrated extraordinary easy and low-cost fabrication of flexible transparent conducting film (TCF) using graphite powder as conducting material. The method is ignoring heating and transfer process which is commonly used in the fabrication of transparent conducting film. Graphite powder is directly and manually deposited onto the transparent plastic by using tube-shaped metal which its round surface was covered with tissue of 2 mm thick. The deposition of graphite powder was performed by circle motion of 20 movements for one coating or one layer. Numerous layers of graphite film were coated on the previous stacked layer to increase TCFs conductivity. Resistivity and transmittance measurement of TCF was performed by four-point probe method and UV-Vis equipment respectively. It is confirmed that graphite TCF achieved the resistivity of 0.98 ohm.cm and transmittance of 60% for one time of coating, while 40 times of coating resulted in the resistivity of 0.17 Ohm.cm and transmittance of 4.6%. Although its transmittance still has a space to be improved higher, our method is very promising for future up scalable production of transparent conducting film owing to its highly easy process and effective cost.

1. Introduction

Transparent Conducting Films (TCFs) hold a crucial key in nowadays developing optoelectronic devices, such as organic solar cell, display, touch screen, Liquid Crystal Display (LCD), Organic Light Emitting Diodes (OLEDs), etc. It is the main gateway where the light passes through and absorbed by the active material of the devices that make carrier generation occurs.

The most widely used Transparent Conducting Film is Indium Tin Oxide (ITO) and fluorine doped tin oxide (FTO), but Indium material scarcity [1], high cost [2], brittleness [3], susceptibility to ion diffusion into active material [4], limited transparency in the near infrared region, and current leakage of FTO fuel the search for cheaper and better alternative of Transparent Conducting Films [5].

Presently, there have been developed numerous TCFs alternatives. Generally, the research was comprised of material used as conducting material and deposition method locating conducting material on the substrate. Among conducting materials that frequently used in the fabrication of transparent conducting films are carbon nanotubes (CNT) [6], graphene [7], nanowire meshes [8], metal grids [9], and ultra-thin metal films [10]. Transparent Conducting Oxide with other material replacing indium is also developed, such as ZnO:Al (AZO) [11] and ZnO:Ga (GZO) [12]. The material cost concern still plays the most important role in choosing the material of transparent conducting material. Selecting the cheapest material is the earliest step in producing a transparent conducting film.
Some deposition methods have been also developed to meet the ideal candidate of commercialized transparent conducting film. Among of them are Metal Organic Chemical Vapor Deposition (MOCVD) [13], Metal Organic Molecular Beam Deposition (MOMBD) [14], CVD (Chemical Vapor Deposition) [15], PECVD (Plasma Enhanced Chemical Vapor Deposition) [16], Solution Deposition [17], Spray Pyrolysis [18] etc. The developed method of deposition still face the problems since easy method can only produce small quantity and sophisticated one suggest high-cost process. Low cost conducting material and easy process of fabrication of transparent conducting film hold the most crucial factor to its up scalable and commercialization production.

Graphite powder is an ideal candidate for conducting material of transparent conducting film due to its abundant availability, low cost, and good electrical and mechanical properties [19]. It is actually a thick layer of graphene which has outstanding optical and electrical properties for the application of transparent conducting material [20-22]. In this research, we proposed graphite powder as conducting material which is deposited on transparent plastic manually without any heating and transfer process. Low cost of conducting material and highly easy deposition process of it are demanded to reduce the cost of transparent conducting material production.

2. Material and Experiment

Graphite powder (Giva utama, Indonesia) with mass of 2 g was manually smeared on the surface of the tube-shaped metal which its surface covered with tissue of 2 mm thickness. The attached graphite powder on the surface of tissue was then deposited on the surface of transparent plastic (Yashica, Indonesia) with the size of 3 x 3 cm² using manual circle motion with 20 movements for one coating time. The process was repeated to increase the conductivity of the transparent conducting film. The coating process was carried out inside of a room with room temperature. Good attachment of graphite powder on the surface of plastic was confirmed by the absence of graphite powder released from the plastic after coating process even with touching activity of hands.

Graphite flexible film was then characterized by using UV-Vis electrometer (Mikropack Brands, NanoCalc 2000, Florida, USA) to investigate its transmittance property. Scanning Electron Microscope (SEM) (JEOL JSM-6360LA, operated at 20 kV) was utilized to observe film morphology. Keithley 224 current source and Keithley 182 voltmeter were used to measure film resistivity by using Four-Point Probe Method. XRD characterization was carried out to get the information about crystallinity pattern of graphite deposited on the transparent plastic. The film thickness was characterized by using an electronic microscope. 2D-porosity characterization was conducted to know how well the graphite powder covered the surface of the plastic homogenously by using an electronic microscope with 40 times of magnificent.

3. Result and Discussion

Figure 1 Confirmed that graphite powder was well attached to the surface of transparent plastic even after touching activity of hands. The film stability was quite good since it still kept the same resistivity and transmittance value after 1 year.
Figure 1. Photo of graphite-based flexible transparent film after 1 time of coating

Figure 2 (a) and (b) revealed that graphite powder was homogeneously deposited on the transparent plastic. The graphite powder was electrostatically attached since the heating process was unused. Graphite powder is in micrometer size. This is supported by XRD characterization (figure 3) which informed the pattern of graphite powder crystallinity exhibiting peaks located at 26.5°, 44.3°, and 54.5° relating with planes (002), (101), and (004), respectively. This film has good crystallinity indicated by enough sharpened peak, the body of peak is not too wide and dominance intensity at 002 plane.

Figure 2. SEM image of (a) surface morphology of graphite-based flexible film. (b) Cross section of graphite-based flexible film after 5 times of coating.
Table 1 informed us the relationship between number of coatings and its transmittance, thickness, 2D-porosity and resistivity. The success of coating graphite powder on the surface of transparent plastic was followed by the investigation of influence of number of coatings to transmittance and resistivity of the film. Both are the most crucial factor of good quality of transparent conducting film. High quality of transparent conducting film was indicated by low resistivity but high transmittance.

Table 1. Relationship between number of coatings and the transmittance, thickness, 2D-porosity and resistivity of graphite-based film

| Number of coatings (times) | Transmittance (%) | Thickness (µm) | Resistivity (Ω.cm) | 2-D Porosity (%) |
|---------------------------|-------------------|---------------|--------------------|------------------|
| 1                         | 60                | 1             | 9.8 x 10^{-1}      | 67               |
| 5                         | 33                | 4             | 8.7 x 10^{-1}      | 59               |
| 10                        | 25                | 6             | 7.8 x 10^{-1}      | 35               |
| 15                        | 16                | 7             | 2.8 x 10^{-1}      | 34               |
| 20                        | 6.5               | 8.5           | 1.5 x 10^{-1}      | 24               |
| 25                        | 6.7               | 9.4           | 1.9 x 10^{-1}      | 24               |
| 30                        | 5.7               | 10            | 4.1 x 10^{-1}      | 18               |
| 35                        | 6                 | 8.4           | 2.5 x 10^{-1}      | 29               |
| 40                        | 4.6               | 8.5           | 1.7 x 10^{-1}      | 21               |

Firstly, we observed the influence of coating number on the film transmittance. We increased the number of coating until 40 times and observing its influence for every 5 times of coating. We found the transmittance of the film decrease from 60% to 6.5% with the increase in the number of coatings of 20 times (Figure 4a). It is predicted that by the increase of the number of coating, more graphite powders were attached to the plastic. The more graphite powder attached to the plastic, the more photon absorbed by the film. Since the film absorbed more photons, it would decrease the transmittance of the film. The decrease of transmittance reached the saturated point in the position of 20 times of coating. The coating of more than 20 times does not significantly influence the film transmittance (Figure 4a). To get the reasons for this phenomenon, we directly investigate the influence of coating numbers on the film thickness. Figure 4b informed us the influence of the coating numbers on the film thickness. The film thickness increased with the increase of the coating numbers.
This is due to more graphite powder attached on the surface of the film. The thickness reached the optimum point by the coating number of 30. Coating more than 30 times does not increase the film thickness anymore (figure 4b). This is what makes the film transmittance not decrease significantly after 20 times of coatings. The film thickness does not increase after coating of more than 30 times. It is inferred that some graphite powder started to be released when the coating process conducted for more than 30 times. Sutisna et al experienced the same experience with the different material. They coated TiO2 particle on the surface of plastic seed and get the optimum thickness of TiO2 particle attached on the surface of plastic seed [24].

The investigation was continued to observe the influence of coating number on the resistivity of the film, another important factor in determining a good quality of a transparent conducting film. Figure 4 (c) guided us to deeply see the influence of it. It clearly informed us the decrease of film resistivity with the increase in coating number. The resistivity started at 0.98 Ohm.cm for 1 time of coating and continuously decrease with the increase in coating number. The decrease of film resistivity reached the lowest point of 20 times of coating with 0.15 Ohm.cm and started to rise after that but went down again after 30 times of coating. This phenomenon remains strange since there is not a clear relationship between film transmittance and thickness especially after 20 times of coating. We started to see another parameter influencing film resistivity and closely explaining this phenomenon. We observed film coverage with graphite powder (2-D porosity of film) and its relationship with coating number. We found that the surface of the plastic increasingly covered by the increase of coating number and relatively become stable after 20 times of coating (figure 4d). Figure 5 shown us clearly the decrease of 2-D porosity with the increase in coating number. Finally, we got a clear relationship between 2-D porosity of the film and its resistivity (Figure 4e). The larger area of the plastic surface covered by the graphite powder, the less resistivity the film posses. The resistivity kept relatively stable after 20 times of coating which has a 2-D porosity of 24%. It is logically in the same of the street condition which has fewer holes would allow the vehicle runs faster on the surface of that street.
Figure 4. Graph of relationship between (a) number of coating and film transmittance (b) number of coatings and film thickness (c) number of coating and film resistivity (d) number of coating and film 2-D porosity and (e) 2-D porosity and film resistivity.
Figure 5. Image of 2-D porosity of graphite-based flexible film (a) one coating (b) 5 times of coating (c) 10 times of coating (d) 15 times of coating (e) 20 times of coating (f) 25 times of coating (g) 30 times of coating (h) 35 times of coating and (i) 40 times

4. Conclusion
We have successfully fabricated graphite-based flexible film by utilizing graphite powder as conducting material. We employed manual deposition method in depositing graphite powder on the surface of the transparent plastic. This film achieved the lowest resistivity of 0.15 ohm for 20 times of coating with the transmittance of 6.5%. This achievement still needs further improvement since the widely used transparent conducting posses $10^{-4}$ Ohm.cm and transmittance higher than 90%. Our research arrived at the conclusion that the most crucial factor in achieving low resistivity and high transmittance of a graphite-based flexible conducting film is its 2-D porosity. The next research was quietly requested in finding the way how to cover the surface of the film homogenously with less 2-D porosity but with least coating number.

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