Rapid Fabrication Approach of Graphene Sensor for Biological Application

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***Abstract*- In this work, we show that sensitive sensing platforms can be derived from the laser irradiation of a non-conducting and flexible material using a laser machine, which can then be used to implement the sensing element of a biosensor to detect and determine the concentration of the target analyte. The aim is to investigate and develop new ways of producing an inexpensive Graphene sensor, implemented as a Radio Frequency (RF) filter for biological application. A Graphene-like material can be produced by the laser irradiation (Laserscribe) of Kapton tape in the form of a filter. The fabrication of this designed filter geometry was achieved by using a laser machine to irradiate the Kapton tape on the chosen substrate (Flame Retardant 4 (FR-4)), by applying the earlier obtained parameters for the fabrication. Several power ratings were used for their fabrication and their conductivity was found to range from 171 x10^{-6} S/mm to 279 x10^{-6} S/mm. The ANSYS (Analysis of System) simulation indicates good results, and options to enhance the material property are also being reviewed. Tests are currently on the way to using a Vector Network Analyser (VNA) to verifying their viability of being employed as the sensing element of a biosensor, thereby leading to their potential application in biological sensing.**

Keywords-Laserscribe; Graphene; Biosensor; Radio Frequency filter; Kapton tape.

I. INTRODUCTION

There is a need in the medical diagnostics for accurate, fast and inexpensive devices, which can be routinely used. In this context, biosensors are considered to provide a viable solution to the problems posed by the contemporary healthcare industry. This is because these sensing devices offer considerable advantages such as: specificity, small size, faster response than laboratory test and cost. These cost-effective, practical, portable, and robust diagnostic devices have received significant interest since there is an important need for bio-devices that can rapidly and precisely analyse biological samples. Diverse ranges of chemicals, biochemical and biological analytes have been detected using sensors made form carbon materials. The electron transfer rate and the performance of these devices are significantly affected by the structure of the carbon material itself. This is due mainly to the electronic states density, and the edge plane site obtainable on the carbon material surface [1][2]. There has been previous work on Laserscribe Graphene Oxide (LSG) like in [1], where Graphene Oxide (GO) film was used to produce LSG electrode, using lightscribe Digital Versatile Disc (DVD) label writing technology, which allows for direct laser writing of the DVD on the GO. Tian et al. fabricated a flexible strain sensor using a light scribed DVD burner to reduce GO film, they achieved a flexible Graphene strain sensor with a low Gauge Factor (GF) for high a deformation application, and a high GF for a low strain sensing application [3].

The shortage of Graphene growth and patterning techniques has become a serious issue affecting its deployment in several specialised applications [3]. Graphene, through synthesis and engineering design, can possess 3-dimensional (3D) structures and porosity, earning them wide range of applications like: composite filter, disposable electronics, energy storage devices and flexible electronics [4]-[6]. In spite of these merits, the current synthesis method of porous Graphene requires high temperature processing of multi-stepped chemical synthesis routes, thereby limiting their widespread commercial applications [4]. Therefore, straightforward synthesis of Graphene based materials in a scalable manner is still an important technological goal in achieving commercialised applications for Graphene material especially in the field of biochemical and biological applications [4]. Glassy carbon has been produced from insulating Polyimide (PI) through pulse ultraviolet laser treatment [4][7], and microsupercapacitor was produced from commercial polymer film using CO_2 infrared laser [4].

However, the technique of laser scribing for the fabrication of an RF low pass filter microstrip transmission line has hitherto not been demonstrated. In this work, we show the production of a microstrip filter made of a Graphene-like material from a non-conducting film by laser reduction technique, which has indicated a vista in the biological sensing application of LSG. Maher et al. used a standard light scribe DVD optical drive to do direct laser reduction of graphite Oxide film to Graphene [8]. Their mechanical strength, electrical conductivity and specific surface area indicated good results, which was then used directly as electrochemical capacitor electrodes, where devices made from these electrodes indicates high energy density values in different electrolytes, while maintaining good electrochemical attributes under high mechanical stress, making them a good contender for high power flexible electronics [8]. All these previous works have shown the viability of an LSG material in biological and electrochemical applications, hence, the need to develop cheaper and easily fabricated means of deriving a suitable sensing platform from LSG material. Since the material for implementing a sensing platform is determined, the next is to look at the shape this sensing platform should take.

There is an increase in demand for microwave systems to meet the emerging telecommunication challenges with respect to cost, size and performance [9]. This work describes the design of a microstrip filter for biological application. Microstrip is an electrical transmission line which can be fabricated using a Printed Circuit Board (PCB) technology, employed to carry microwave frequency signal [9]. It comprises a conducting strip separated form a ground plane by a dielectric substrate and it much cheaper than the traditional waveguide technology [9]. This microstrip will be designed in the form of a filter. Since filters are crucial components in microwave communication systems and RF circuits, they are used to either combine or separate the selected frequencies in the band, to fulfil the required specification [10].

Microstrip line is a good technique for filter design due to its benefits of low cost, light weight, compact size, planar structure and its ease of integration with other component on a single board [11]. By extension microwave filters can be divided in to two different types; the lumped element, consisting of capacitors and inductors and the distributed element which used lengths and widths of transmission lines to create their inductance or capacitance value [11][12], resulting in an Inductor Capacitor (LC) ladder type stepped impedance Low Pass (LP) microstrip Filter if a LPF was the design intension [13]. The structure of this work is as follows. Section II deals with the microstrip mathematical design, while Section III deals with the experimental techniques. In Section IV, the results were illustrated, and the Section V handles the discussion and conclusion.

II. DESIGN METHODS

A. Microstrip Transmission Line Design

Several different equations have been developed for use in determining the characteristic impedance for microstrip design. Probably the most convenient are the following which are stated to be accurate to within about 1% [13]. A simple but accurate equation for microstrip characteristic Impedance is shown below with Figure 1 being the microstrip arrangement [14]:

$$Z0 = \frac{60}{\sqrt{\varepsilon eff}}\left\{log\left(8\frac{h}{w} + \frac{w}{4h}\right)\right\} \quad (1)$$

where the effective dielectric constant is given by (2) as:

$$\varepsilon eff = \frac{\varepsilon r+1}{2} + \frac{\varepsilon r-1}{2}\left\{\left(1+12\frac{h}{2w}\right)^{-1/2} + 0.04\left(1-\frac{w}{h}\right)^{2}\right\} \quad (2)$$

while (3) and (4) are for when $w/h \geq 1$

$$Z0 = \frac{120\pi}{\sqrt{\varepsilon eff\left\{\frac{w}{h}+1.393+0.667log\left(\frac{w}{h}+1.444\right)\right\}}} \quad (3)$$

$where:$

$$\varepsilon eff = \frac{\varepsilon r+1}{2} + \frac{\varepsilon r-1}{2}\left\{\left(1+12\frac{h}{2w}\right)^{-1/2}\right\} \quad (4)$$

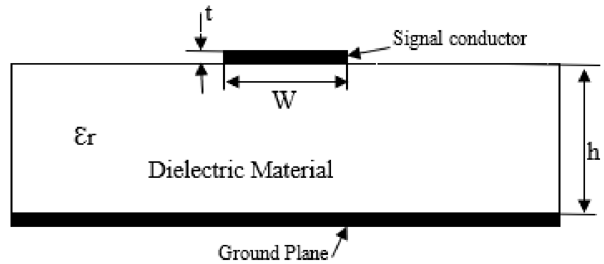

Figure 1. Microstrip Transmission line arrangement

For the purpose of this work, the design was based on the second sets of conditions where $w/h \geq 1$, hence equations (3) and (4) had to be employed.

B. Microstrip Transmission Line Design for A Single Strip Case

From equations (3) and (4), the width of the proposed microstrip line was then calculated since for a wide microstrip (i.e. width=W), nearly all of the electric field lines will be focused between the metal planes, similar to the case of a parallel plate capacitor. On the other hand, for narrow W the electric field lines will be about equally divided between the air and the board dielectric [13].

To get the effective dielectric constant, (4) would have been used, however, there are two unknowns, as a result, the formula that gives an approximate value of the characteristics impedance was then used, which is given as shown in (5) below [12][15].

$$Z0 = \frac{87}{\sqrt{\varepsilon r}+1.41}\left\{log\left(\frac{7.48h}{w-1.25T}\right)\right\} \quad (5)$$

From which:

$$w = \frac{7.48h}{e^{\left(Z0\frac{\sqrt{\varepsilon r+1.41}}{87}\right)}} + (1.25t) \quad (6)$$

For the purpose of this work, an FR-4 dielectric material was employed, with a dielectric constant ranging from 4 to 4.47 [12][16]. From the pre-test samples, the thickness T was found to be 4.8532 mil. The Material used for this work was, an FR-4 board of dielectric thickness of 10mil. The characteristic impedance of choice for the material is 50Ω. From the information above, the following was deduced:

T=4.8532mil

H=10mil

Z0=50 Ω εr=4.4

By putting all these parameters in equation (6) above the width of the material can be calculated as shown below:

$$w = \frac{7.48\times 10}{e^{50\left(\frac{\sqrt{4.4+1.41}}{87}\right)}} + (1.25\times 4.8532)$$

w=24.7852mil≈25mil

w=0.635mm

This value is close to the value measured using a Mitutoyo stylus surface roughness measuring tester (Surftest SJ-410) where the thickness value was measured to be 0.4835mm.

C. Microstrip Transmission Line Design for A Multiple Strip Case

For this case, the strip was made up of several single strips (for this work, several single strips were used), as such to calculate the width of this strip, it would have been the addition of the width of the individual single strips and the resultant sum becomes the width of this strip (i.e. a multiple strip). However, this is not so because during the production of this material it was observed that the machine always makes overlapping tracks on the Kapton tape which means the width of the material produce would not be brought about as expected. As a result, the width of this material was not calculated but was measured using the Surftest SJ-410, from this measurement the width was obtained as 1.75875 mm which is equivalent to70 mil. In a bid to obtain the accurate results for each of the slope of the results measured from the resistance of the materials along its length, an analytical method of determining the slope of the straight line was used. To achieve this, the method of least squares was employed.

III. EXPERIMENTAL TECHNIQUE

The mathematical design of the microstrip was done in the previous section where the calculation (Mathematical formulation) of the width and expected thickness was made. The implementation of this design was achieved using the laser scribe technique for making GO. This technique of GO production is one of the do-it-yourself methods of producing Graphene. This technique has exposed one to a new way of making a Graphene Oxide or rather a Graphene Oxide-like material. To achieve this task several experiments were conducted, and the materials that were initially used were; Kapton tape, silver paint, plywood (substrate), digital multimeter and laser machine (Laserscript HPC laser) with laser spot size of 0.2 mm and a vertical scanning position. There are two basic categories of material, from which eight samples of these materials with varying parameters were produced. The two categories of samples were; the Multiple-Strip (MS) line and the Single Strip (SS) line. The two main parameters that were varied are the speed at which the laser engraves on the Kapton tape and the thickness of the width of the strip line.

The MS line sample has four set of samples, where each sample is made up of five materials with varying laser power settings ranging from 26% to 34% in increment of 2% (this was determine base on test carried out on the pre-test samples to ascertain the power settings that would produce a material with the best property). This fabrication principles also applies to the single strip-line, it has the same fabrication principles and the same parameters as its multiple-strip counterpart but differ by the number of strip line on it (in its own case it is just a single strip line). For the multiple strip, the two main parameters which were the speed at which the laser engraves on the Kapton tape and the width of the microstrip, these were chosen and set. These parameters changes from sample to sample to produce the four sets of samples for each category of sample. The same principles apply to the single strip samples. For both the MS line and the SS line the samples main parameters are shown in Table I below.

TABLE I. SETTINGS USED TO PRODUCE THE MICROSTRIP

parameter	Sample 1	Sample 2	Sample 3	Sample 4
Speed (mm/s)	300	300	400	400
Width (µm)	300	350	300	350
% power	26, 28, 30, 32, 34.	26, 28, 30, 32, 34.	26, 28, 30, 32, 34.	26, 28, 30, 32, 34.

These sets of settings were employed for both the multiple strip line and the single strip line, which resulted in samples amounting to eight in total. To achieve the design of these samples several steps were followed; Firstly, after the mathematical design, some Kapton tapes were then laid (taped) unto the surface of FR-4 material. After which, the settings for the category of interest was done on the system (computer). Then, the settings for the material of interest was also done on the system before transferring these settings to a flash drive, and on to the laser machine. The sample obtained from the first step above was then introduced into the working area of the laser machine to begin the irradiation process. The same procedure was repeated from the first step to the fourth step for all the two categories to produce the entire forty materials. From these samples produced, several tests were carried out on them using a digital multimeter to obtain necessary resistance values from the resulting materials (Graphene Oxide-like). The reason for carrying out this test is to determine the resistivity of the resultant material to ascertain the viability of its being employed as a material that has the required property for use as the sensing element of the proposed biosensing platform. The two categories of samples (the multiple strip and the single strip) were tested to explore which material from any of the two sets of materials (twenty different materials for the multiple strip and twenty for the single strip) is viable and results were taken.

The results of these tests carried out were recorded. From these results, several plots were also done, and with further manipulations it led to the determination of the material parameters that is optimum and suitable for the design of the material of interest. In order to determine the suitable resistivity of the materials produced, the areas of each material needed to be determined, and to achieve this task, the thickness and the width of the individual materials were measured using the Mitutoyo stylus surface roughness measuring tester (Surftest SJ-410), with this device the measurement were done to the nearest micron. This technique was used to fabricate a 5th order LPF microstrip LSG on a PCB substrate for use in the intended application. Figure 2 below gives the steps by step process of fabricating the LSG microstrip LPF, while Figure 3 shows the fabrication machine and the filter during fabrication.

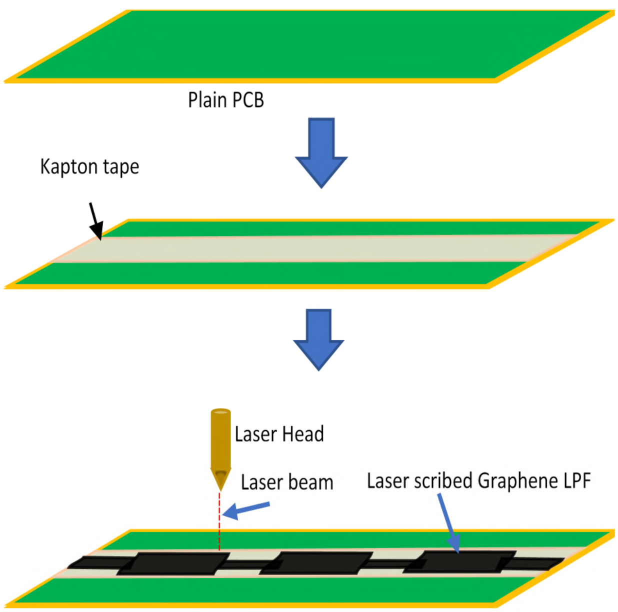

Figure 2. Laser scribed Graphene fabrication process

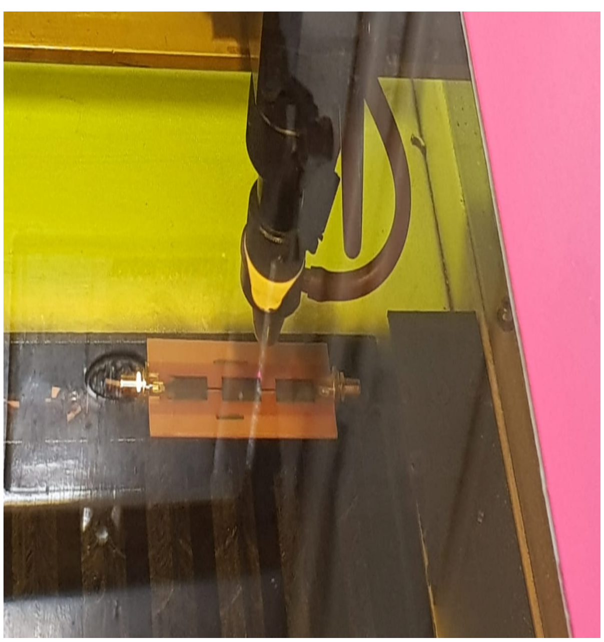

Figure 3. Microstrip LPF Fabrication

IV. SAMPLE RESULTS

The results of the test carried out for the different parameters of the different materials developed, were recorded as measured. From the results obtained it was observed that certain resistance values do not increase linearly with increase in length (i.e. they are irregular). As such, these give an idea of which material, from this set of samples that might possess the needed property for the intended usage. From these results, plots were made to determine the slope which is the resistance per unit length of the produced material. A typical slope for these results is as shown in Figure 4 below. The same procedure was followed to obtain results of the single strip line, using the same equipment as used for the multiple strip line above, results were obtained and recorded as shown below. From these results, plots were also made to determine the slope which is the resistance per unit length of the produced material. A slope typical to these results is as shown in Figure 5 below.

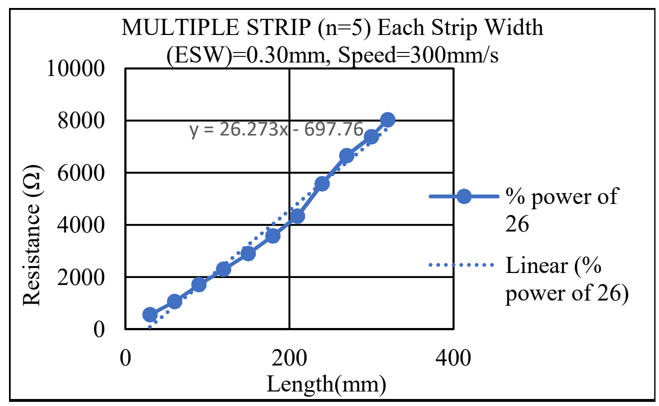

Figure 4. Resistance against length for a multiple strip line material.

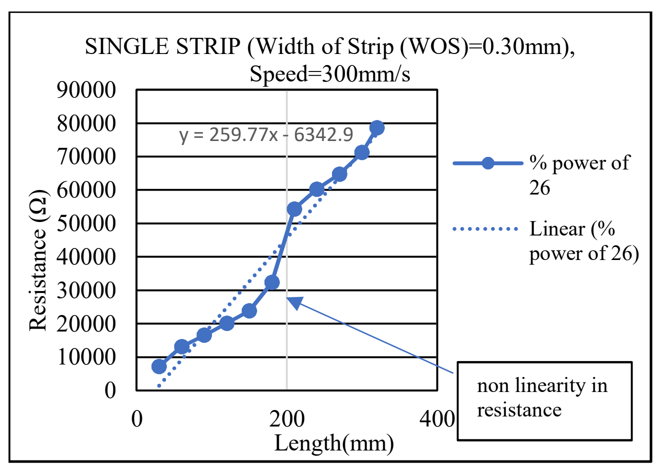

Figure 5. Resistance against length for a typical single strip line material.

From the above results, the resistance per unit length was plotted against the percentage power in both categories for all the strips. The results are as shown in Figures 6a and 6b. From these plots, it has become obvious that there is more discontinuity associated with the SS materials. These

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materials and the percentage powers at which these discontinuities occurred become evident from the plots. From the plot it can be observe that certain points were enlarged, this highlights the materials with observed abnormalities and the corresponding percentage powers at which they were fabricated. These plots have also helped in narrowing down the material with the needed property from either category. As can be seen from the results shown below in Figure 6a and 6b, it can be observed that the resistance per unit length (on the vertical axis) for the single strip material is about ten times that of the Multiple-Strip (MS) material. More so, the number of defective materials as indicated with bold black dots are more prevalent in the single strip materials than its multiple strip counterpart. This is an indication that the multiple strip material has shown promising characteristics to make it a viable option for its design scheme to be implemented for the designing of the microstrip transmission line, which will in turn serve as the sensing element of the biological sensor whose efficacy is dependent on the resistivity of the material.

In a bid to determine the most suitable fabrication scheme for this method of producing this laser scribed Graphene Oxide material, further processing of the results was done which resulted in the plots of Figure 6c and 6d below. These results show typical plot for both categories, from which useful data about the material and the method of making the material can be obtained. ANSYS simulation was done using the conductivity value derived from the test results and the results indicated that the fabricated filter's response was similar to the theoretical lowpass filter as shown in Figure 7.

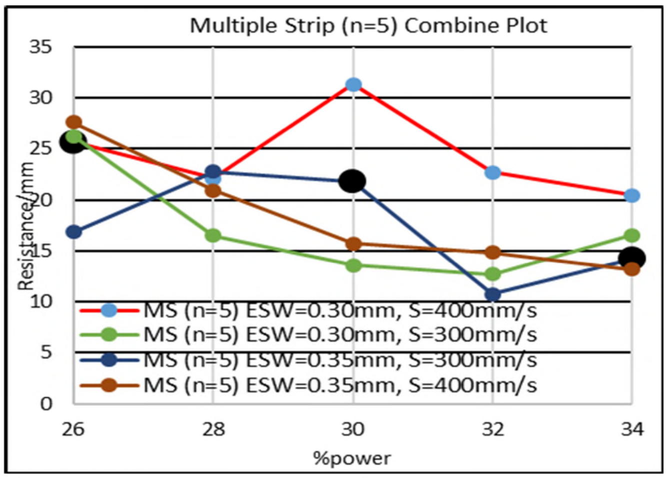

Figure 6a. Resistance/ mm of multiple strip category with black dots indicating defective material

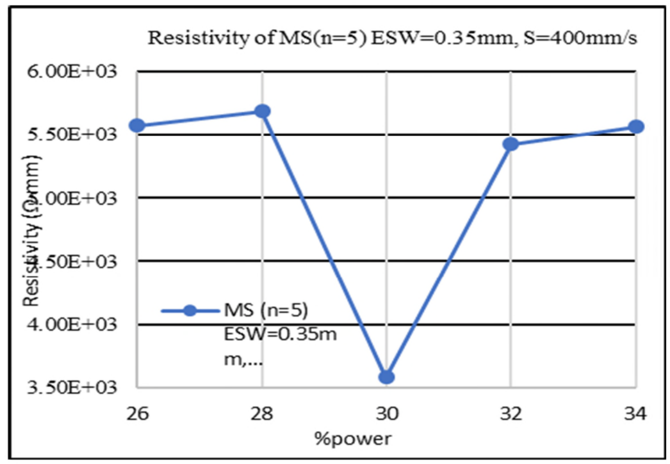

Figure 6c. Resistivity of the multiple strip category with estimated strip width (ESW) of 0.35mm.

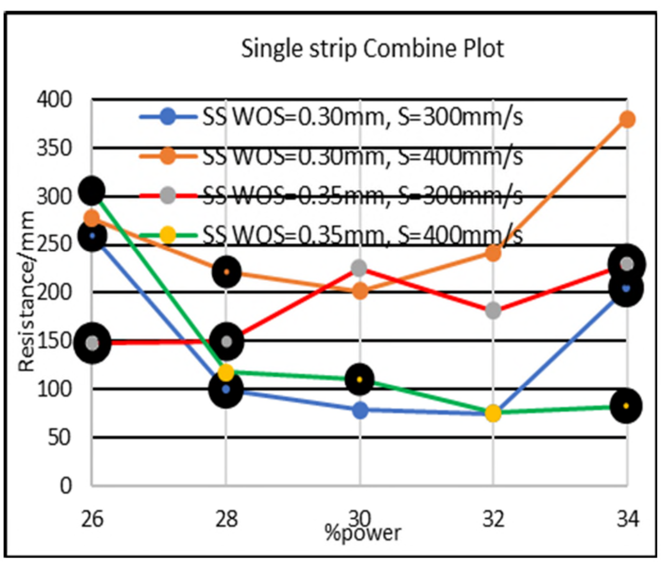

Figure 6b. Resistance/mm of single strip category with black dots indicating defective material

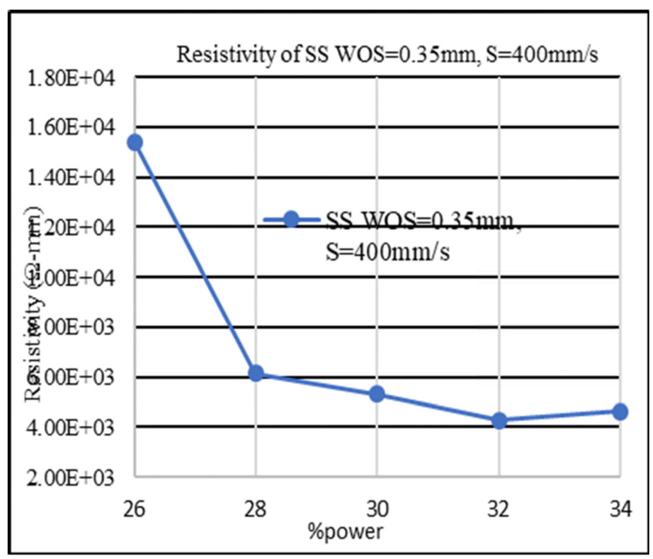

Figure 6d. Resistivity of the single strip category with width of strip (WOS) of 0.35mm

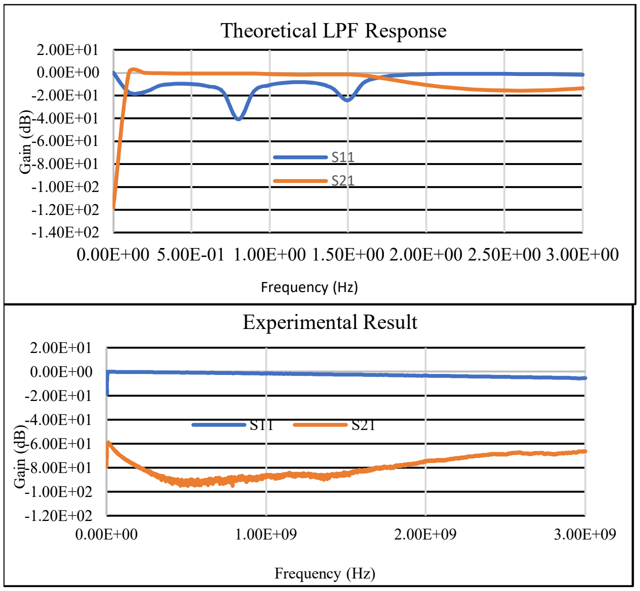

Figure 7. Experimental and Theoretical Results of a Microstrip LPF

Haven produced the 5th order LPF using the MS line technique. VNA was used to measure its S-parameters, to determine suitability for the intended application. From the observed results shown in Figure 7, there is a difference between the expected and the experimental/simulation response of a microstrip LPF, thereby indicating that more fine-tuning of the fabrication parameters is necessary.

V. CONCLUSION AND FUTURE WORK

The aim of this work was to develop new ways of producing a Graphene sensor, starting with cheap rapid prototyping of implementing an inexpensive and flexible electronic material that is easily reproduceable, with high conductivity, and can be deployed for use as a sensor for biological application. The results indicated that the multiple strip line material has a promising conductivity value ranging from 171 x10^{-6} S/mm to 279 x10^{-6} S/mm, signalling its viability for the proposed application. From these results, it can be concluded that this LSG can also be a fine application in various sensing platforms where light weight and flexible conductor is desired. The results of the microstrip LPF implementation indicated the need to fine tune fabrication parameters so as to improve the filter response. The future work includes fine tuning the fabrication parameters and the use as biological sensor. Then, testing the device with a mouse IgG needs to be done, to show its applicability as a biological sensor before moving on to more sensitive analytes like Prostate-specific antigen (PSA). To achieve that, in this work, both a Chebyshev type 5th and 7th order low pass and bandpass filters are to be implemented as the sensing surface using the insertion loss method.

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