CHAPTER 6: GRAPHENE OXIDE-BASED BROADBAND POLARIZER

6.1 Introduction

In this chapter, the fabrication of the polymer waveguide is outlined in details. The waveguide is then drop-casted with graphene oxide (GO) solution to make it a broadband polarizer. A experimental setup is proposed to evaluate the performance of the GO-coated waveguide as a efficient light polarizer. The underlying principles of the strongly polarization dependent propagation loss demonstrated are studied.

6.2 Fabrication of Graphene Oxide-based Polymer Waveguide

A CR-39 polymer sheet with a thickness of 0.5 mm and a refractive index of 1.486, measured at 1550 nm with a Sairon Technology SPA-4000 Prism Coupler (Figure 6.1), is used as the substrate for the polymer waveguide. The optical characteristics of CR-39 resemble those of crown glass and is commonly used in the manufacture of plastic lenses. SU-8 polymer with a refractive index of 1.569 measured also at 1550 nm in a Sairon Technology SPA-4000 Prism Coupler is spin-coated onto the CR-39 sheet and patterned using the contact photolithography technique, forming the core of a channel waveguide structure. The measured height of the channel waveguide was 5.0 (± 0.1) μm and its width ranges between 10 and 15 μm over several different samples, with waveguide width variation of less than 0.2 μm over the length of a given waveguide section.

Figure 6.1: Sairon Technology SPA-4000 Prism Coupler available at the Class 10K Cleanroom of Photonics Research Centre, University of Malaya.

0.5 μl of GO solution containing GO flakes is carefully dispensed onto the waveguide channel using a micropipette and then allowed to dry under ambient conditions before the next GO drop is applied in the same position. Surface mapping of the GO-coated polymer channel waveguide is carried out using a DEKTAK D150 surface profiler (Figure 6.2). In the work here, the sample is covered by NOA-65 UV-sensitive resin (Norland Co. Ltd.) which has a refractive index of 1.52, as an immediate overcladding of the channel waveguide and the GO covered regions. This encapsulation provides both mechanical protection. A major reduction in the susceptibility of the deposited GO to the possible effects of atmospheric water vapour is anticipated. The GO coated samples are then diced and polished into 1 cm long waveguide channels.

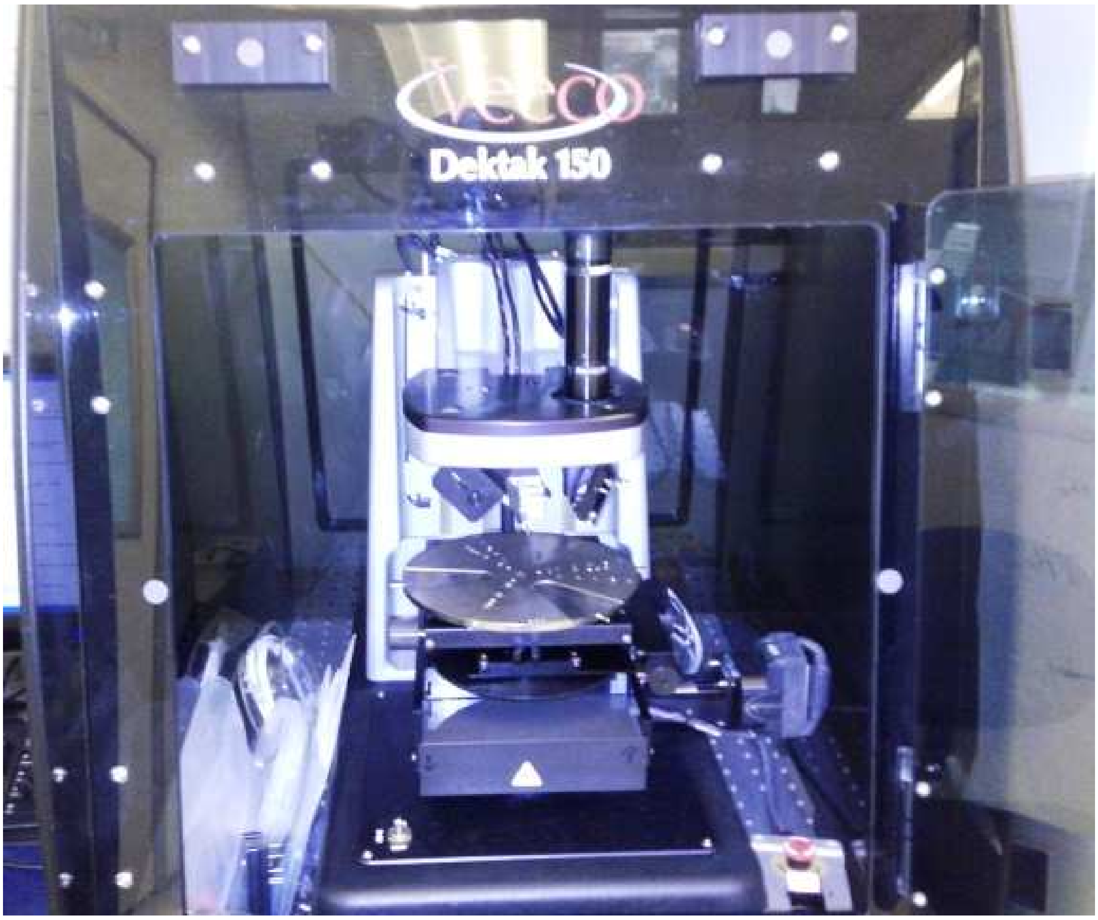

Figure 6.2: DEKTAK D150 surface profiler at the Class 10K Yellow Room of Photonics Research Centre, University of Malaya.

The fabrication process is summarized as shown in Figure 6.3.

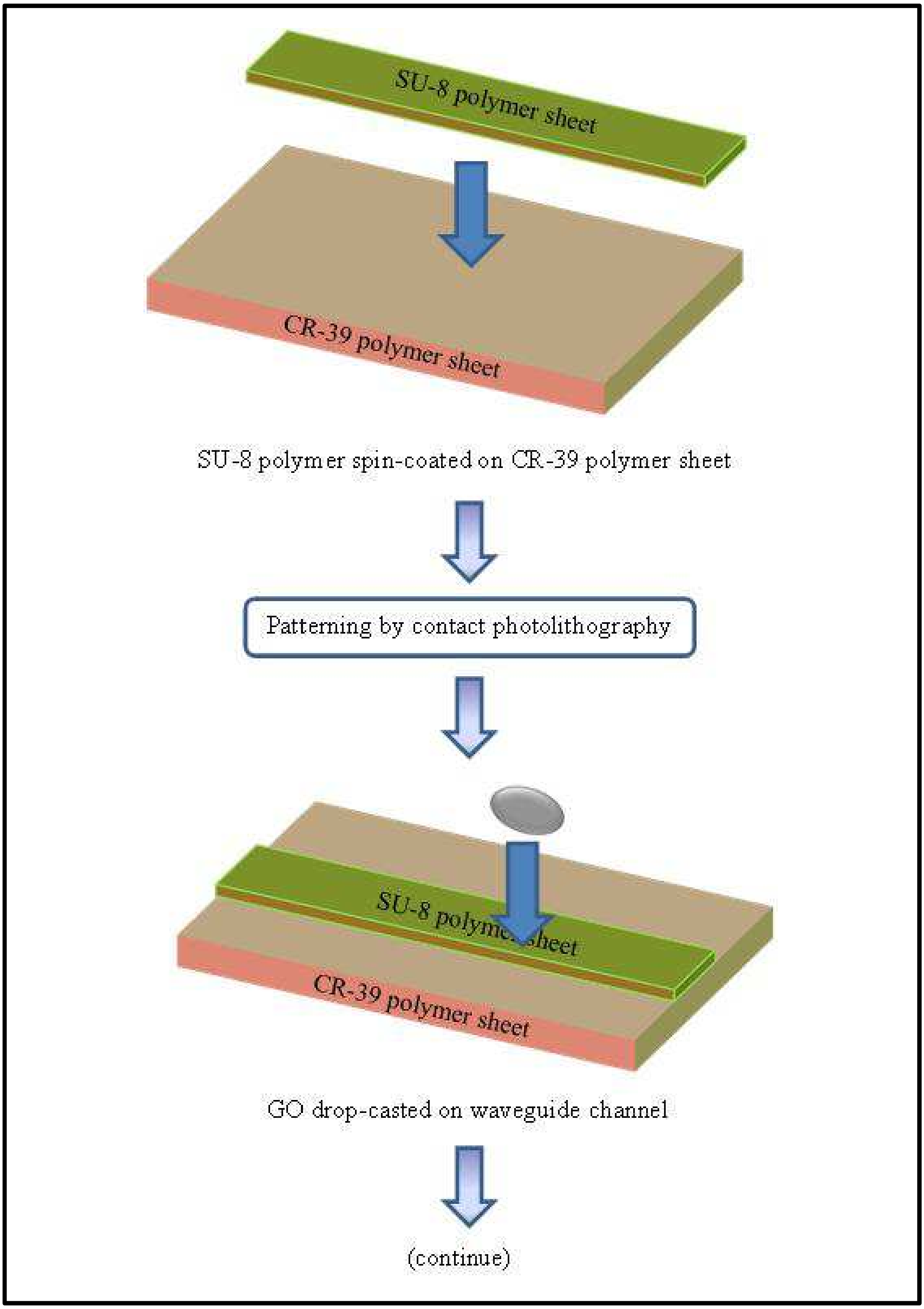

Figure 6.3: Graphene oxide-based polymer waveguide fabrication process (to be continued).

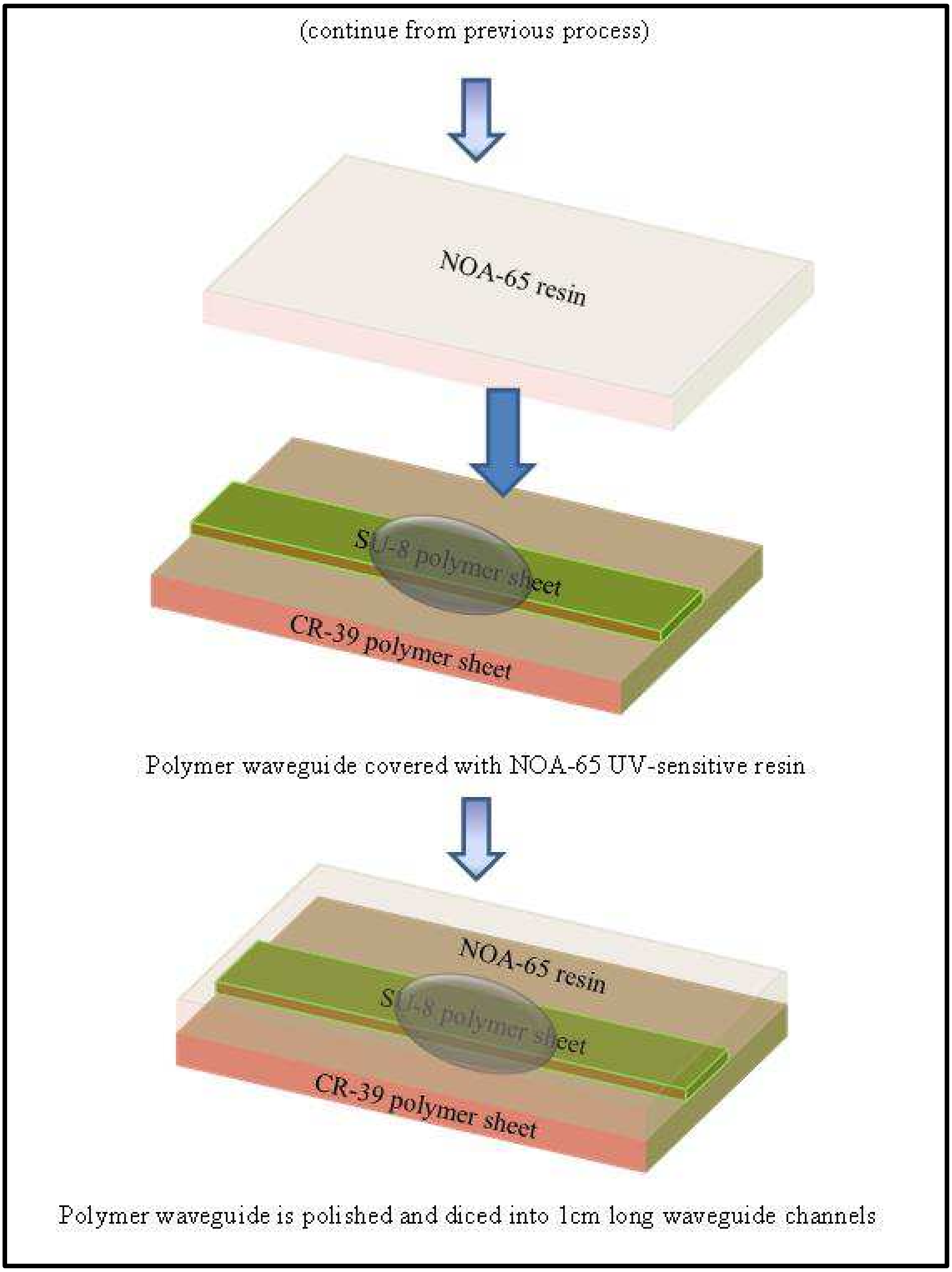

Figure 6.3: Graphene oxide-based polymer waveguide fabrication process (continue from previous section).

6.3 Physical Properties of the Graphene Oxide-based Polymer Waveguide

GO multi-layer is deposited on to the polymer waveguide channel using the drop-casting method, as described in Section 6.2. As a progressively larger number of GO solution drops are applied, the GO film becomes thicker, as shown in Figure 6.4, where the transparency of the GO coating decreases while the coating diameter remains about the same.

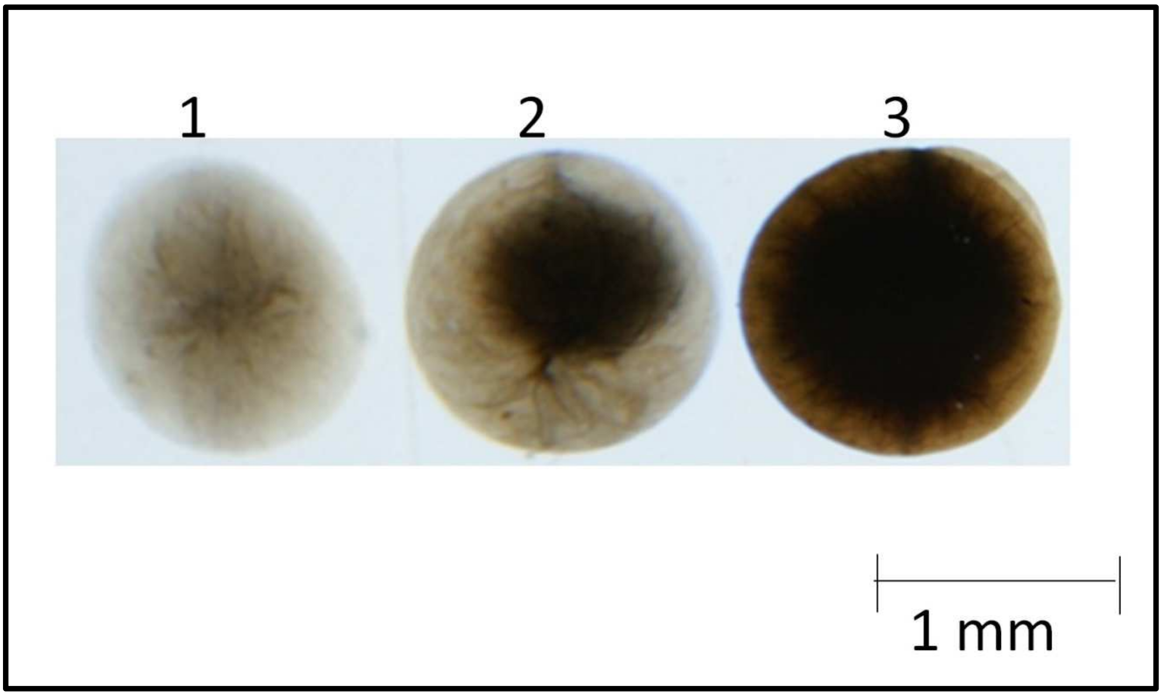

Figure 6.4: Drop-cast GO coating with different drops of solution.

The surface morphology of the channel waveguide coated with a single drop of deposited GO solution is shown in Figure 6.5 (a). The average thickness of the GO film for each solution drop-casting step is ~0.5 μm. It can be seen that the GO film showed similar height coverage, both on the channel waveguide and its surroundings. This coating behaviour has implications for the overall effect of the number of GO solution drops applied during the drop-casting process and the extinction ratio of the proposed waveguide polarizer, as will be discussed in the following section. Figure 6.5 (b) shows a Scanning Electron Microscope (SEM) image of a GO-coated waveguide channel. This figure indicates that the drop-cast GO forms a continuous layer across the coating region. Wedge-shaped air gaps are also observed on either side of the waveguide channel sidewall, indicating the formation of the GO film on top of the waveguide channel during the droplet drying process before the layer formed has “collapsed” onto the waveguide channel, at the end of the drying process. This behaviour also explains the elevated ridges observed on the GO film in Figure 6 (a), which are wrinkles resulting from local folding of the GO film during “collapse”.

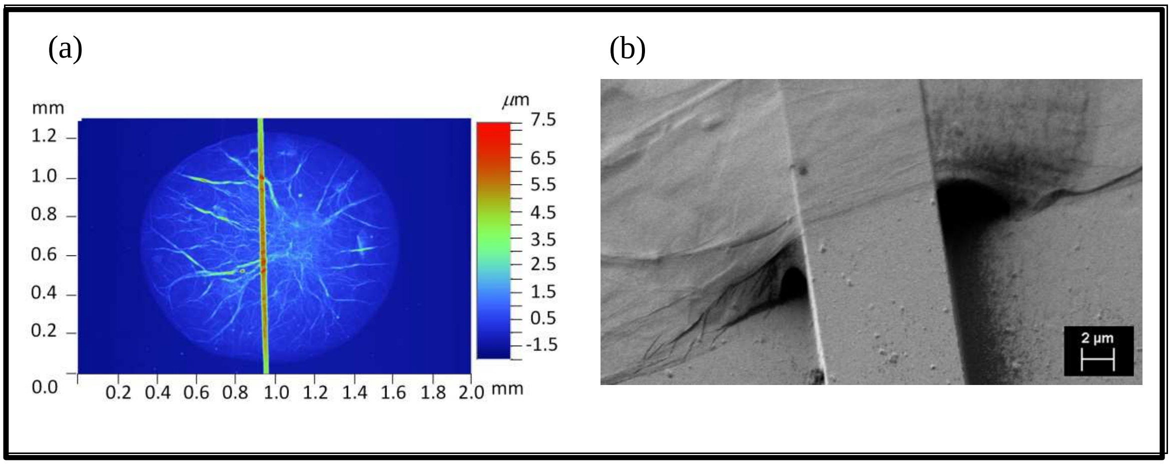

Figure 6.5: (a) Surface mapping of drop-cast GO coating on polymer waveguide channel. (b) SEM image of a GO-coated waveguide channel. Air gaps are observed between the sidewalls of the waveguide and the GO film.

A close-up examination of the GO film formed with 2 drops of deposited GO solutions shows an orderly layered GO stack, indicating the formation of GO ‘paper’ (shown in Chapter 5, Sub-section 5.2.1, Figure 5.10). The spacing between the GO layers is calculated from the XRD result (shown in Section 5.2.1 of Chapter 5), and is about 1 nm. This interlayer spacing indicates the presence of a large number of water molecules intercalated in the GO film, as reported by Lerf *et. al*. [1] and Buchsteiner *et. al*. [2]. No obvious gap is observed between the first and second depositions of the GO film. All these features indicate a clear distinction between the presently considered GO waveguide polarizer and the graphene-based waveguide polarizers described previously [3, 4], since the number of GO layers involved is much larger than the number of graphene layers required to support plasmonic wave propagation. The GO multi-layer of the present work, in contrast, may be characterised as an anisotropically lossy dielectric overlay. It has a sufficiently large refractive index value to produce a significant redistribution of the modal fields compared with the modal fields of the uncoated polymer waveguide.

6.4 Characterization of the Graphene Oxide-based Polymer Waveguide

Fibre butt-coupling technique is used to measure the insertion loss of both the TE- and TM-polarized modes of the GO-coated waveguide channels, as shown in Figure 6.6. Guided light polarization in the launch fibre is controlled using a fibre polarization controller, and the polarization state of the GO polarizer output is measured in free space using a polarimeter (Thorlabs PAX 5710) before the output is fibre-coupled, in order to measure the insertion loss. Although the SU-8 waveguide core cross-section is large enough for several characteristic modes to be available for both (quasi-) TE- and (quasi-) TM-modes, the alignment of the set-up guaranteed that, to a good approximation, only the fundamental guided mode for each polarization is launched. In addition, extra care in the handling of the launch fibre is required, since the linear polarization state could easily be scrambled by small movements or vibration. The polarization-dependent loss of the uncoated polymer waveguide is measured first and is found to be lower than 0.5 dB, limited by the performance of the measurement setup. For measurements across the broad frequency range, fibres with different cut-off wavelengths are used, in order to maintain the launching polarization state and single waveguide mode excitation conditions.

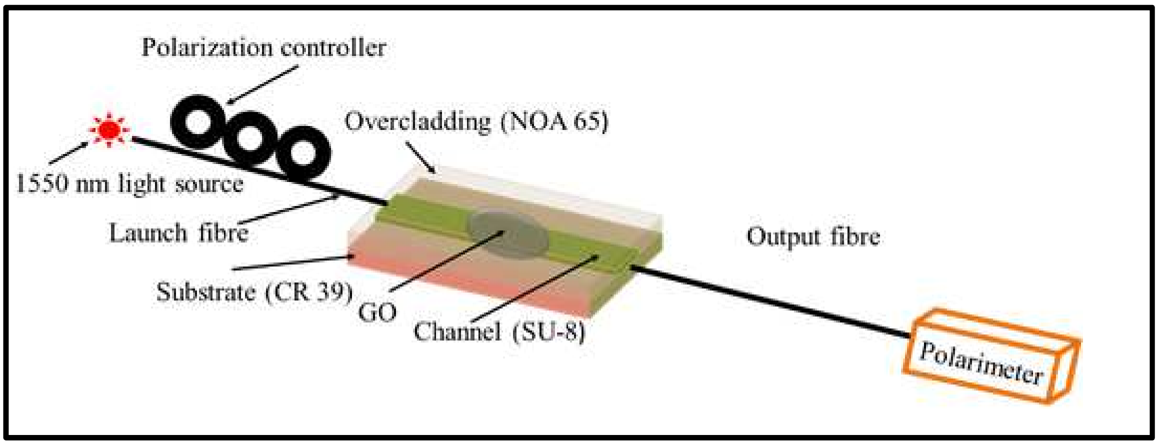

Figure 6.6: Schematic diagram of the experimental setup for the characterization of the GO-based polymer waveguide.

6.5 Performance of the Graphene Oxide-based Polymer Waveguide as a Broadband Polarizer

The performance of the GO waveguide polarizer is measured and the results are discussed. The experimental setup is shown in Figure 6.7. Figure 6.8 shows a polar plot of the measured output light from the GO waveguide polarizer, as a function of the input polarization. The output of the GO waveguide polarizer is characteristically highly TM-polarized - which is similar to the observation reported by Kim *et. al.* [5] and indicates that the waveguide modal distribution changes substantially in the GO coated waveguide section.

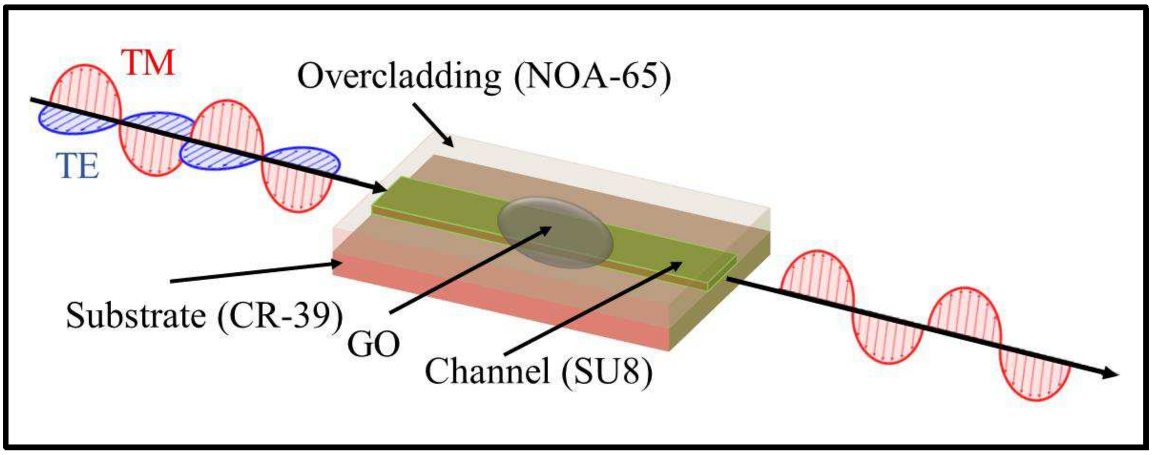

Figure 6.7: Graphene oxide-based waveguide polarizer.

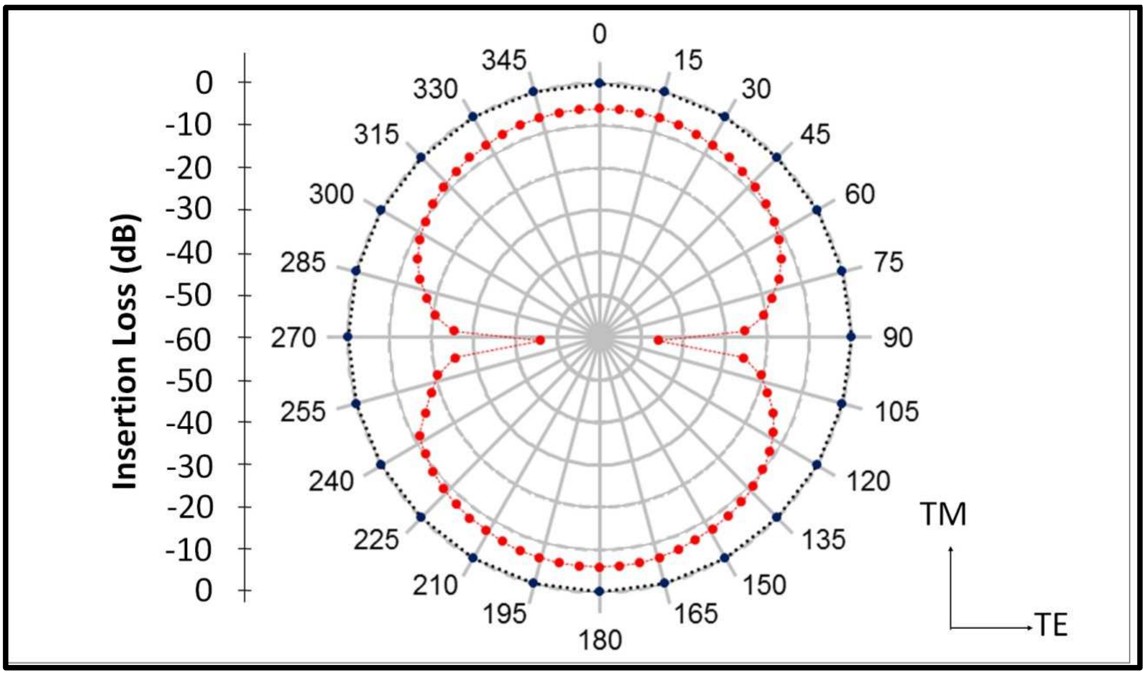

Figure 6.8: Polar plot of the GO waveguide polarizer measured at 1550 nm. The insertion loss is lowest when the incident light is highly TM-polarized, indicating a TM-pass waveguide polarizer.

Figure 6.9 shows the insertion loss for the TM- and TE-polarized modes, for different GO film thicknesses determined by the number of GO solution drops applied during drop-casting. Both values are measurement averages for wavelengths in the range between 1530 and 1630 nm. The total variation of the extinction ratio over this wavelength range is 4.5 dB. The insertion loss for the TM mode increased with the number of drops of GO solution applied and it becomes constant after deposition of three GO drops, at approximately 6.5 dB. As the number of GO drops applied increases, there is a correspondingly rapid increase in the TE-mode propagation loss, which continues to increase steadily and reaches a maximum value for coating with five GO drops, corresponding to a GO film thickness of 2.5 μm.

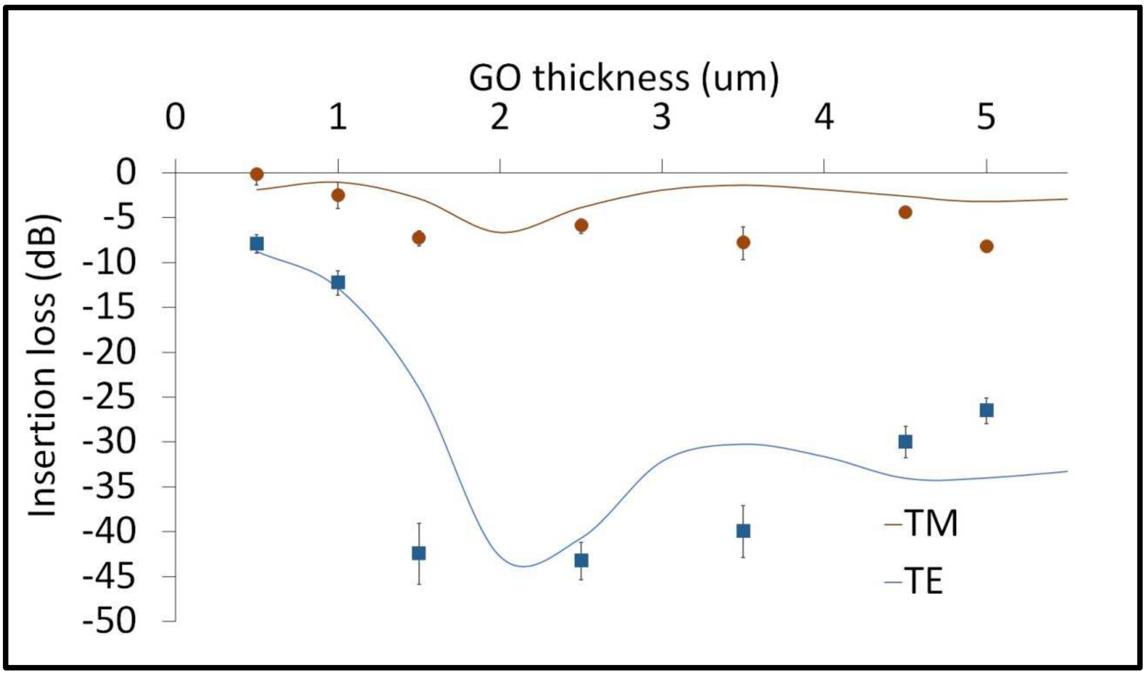

Figure 6.9: Insertion loss of TE- and TM-polarized light of waveguide polarizers coated with different number of GO solution drops corresponding to different GO film thickness. Both values are measurement averages in the 1530 to 1630 nm wavelength range.

The efficiency of the coupling of light into the drop-cast GO multi-layer is likely to be dependent on the thickness of the GO coating on the top channel waveguide surface. At the start of the drop-casting, the initial GO coating thickness is small and it can only support a small part of the total propagating modal power. In this case, a large fraction of the modal power for both TE- and TM-polarized waves remains in the polymer waveguide. As additional drops of GO solution are applied, the GO coating thickness increases and experimental observations indicate that a point is reached where the mode coupling between the waveguide and the GO multi-layer is most efficient. This point is characterized by a sharp increase in the TE mode propagation loss (GO film thickness larger than 1.5 μm), as shown in Figure 6.9. At the same point, the TM mode shows a partial but significant redistribution of modal power from the polymer core into the GO coating.

The TM mode propagation is verified by observing the polarization of the light leaking out from the top surface of the GO coated channel waveguide when a 650 nm wavelength laser was coupled into the channel waveguide, as shown in Figure 6.10. Using a polarization analyser placed between the waveguide and the microscope, the light leaking out from the top surface of the waveguide was found to be in the *p*-polarization state (i.e. parallel to the plane of incidence – the plane defined by the waveguide axis and the direction of the microscope observation). This measurement indicates that TM-polarized light is coupled into and propagates partially in the GO film with relatively small propagation losses, while TE-polarized light experiences a much larger propagation loss in the GO film.

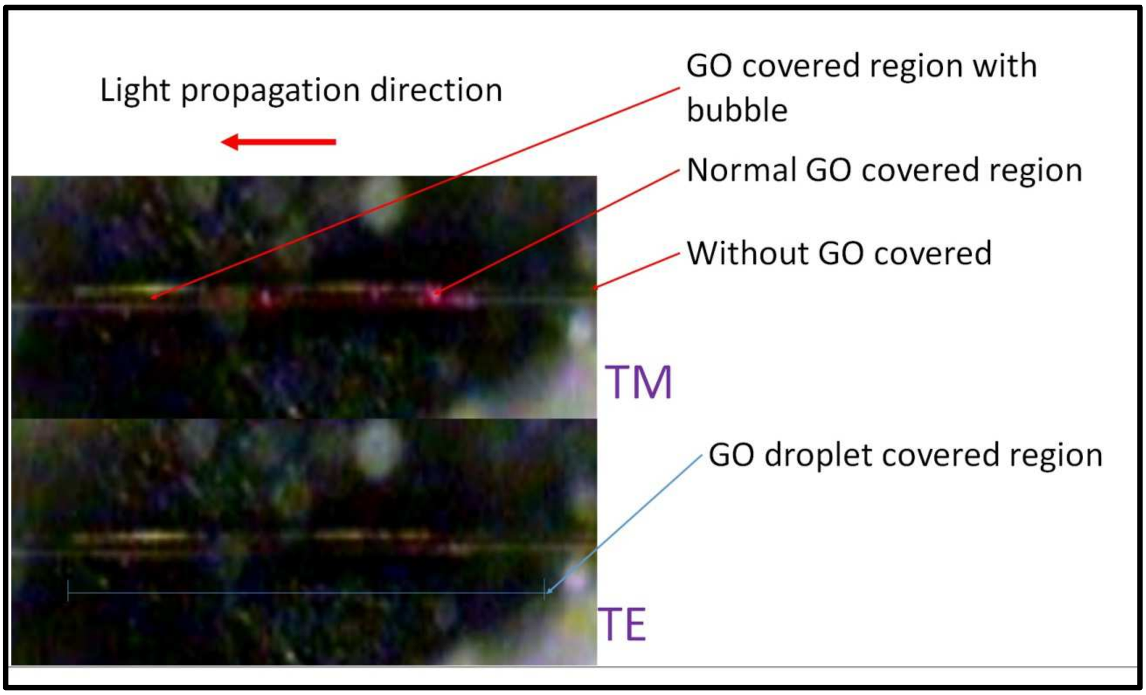

Figure 6.10: Top view of GO coated waveguide channel with TM- and TE- polarized light (650 nm) propagating through the waveguide. There was no observable scattering in the case of TE-polarized light, while the observed scattering in the case of TM-polarized light propagation was highly *p*-polarized.

Addition of more GO solution drops does not change the size of the drop-casting area and therefore it should increase the number of GO layers that is observed as the film thickness increases. The addition of further drops of the GO solution beyond a certain point (typically three solution drops) does not measurably increase the propagation loss of the TM-mode, since the additional layers of GO coating are progressively further from the waveguide-GO interface and therefore interact less strongly with the light propagating in the channel waveguide.

The polarization extinction ratio for wavelengths ranging from 650 nm to 1640 nm has been measured and is shown in Figure 6.11. It shows a gradual decrease in value towards shorter wavelengths. The extinction ratios at wavelengths of 1590 nm, 1310 nm, 980 nm and 650 nm were found to be 40.0 dB, 25.0 dB, 16.0 dB and 8.5 dB, respectively. Though steps have been taken to ensure only fundamental mode propagation in the GO waveguide polarizer, it is believed that partial excitation of higher order modes of the waveguide at shorter wavelengths is quite probable and that their presence would reduce the effective strength of the interaction between the polymer waveguide and GO film, resulting in a reduced extinction ratio between the TE- and TM-polarized modes. Nevertheless, an extinction ratio of more than 20 dB has been measured over the entire wavelength range from 1250 nm to 1640 nm. The extinction ratio for wavelengths longer than 1640 nm was not measured, due to the unavailability of a suitable laser source.

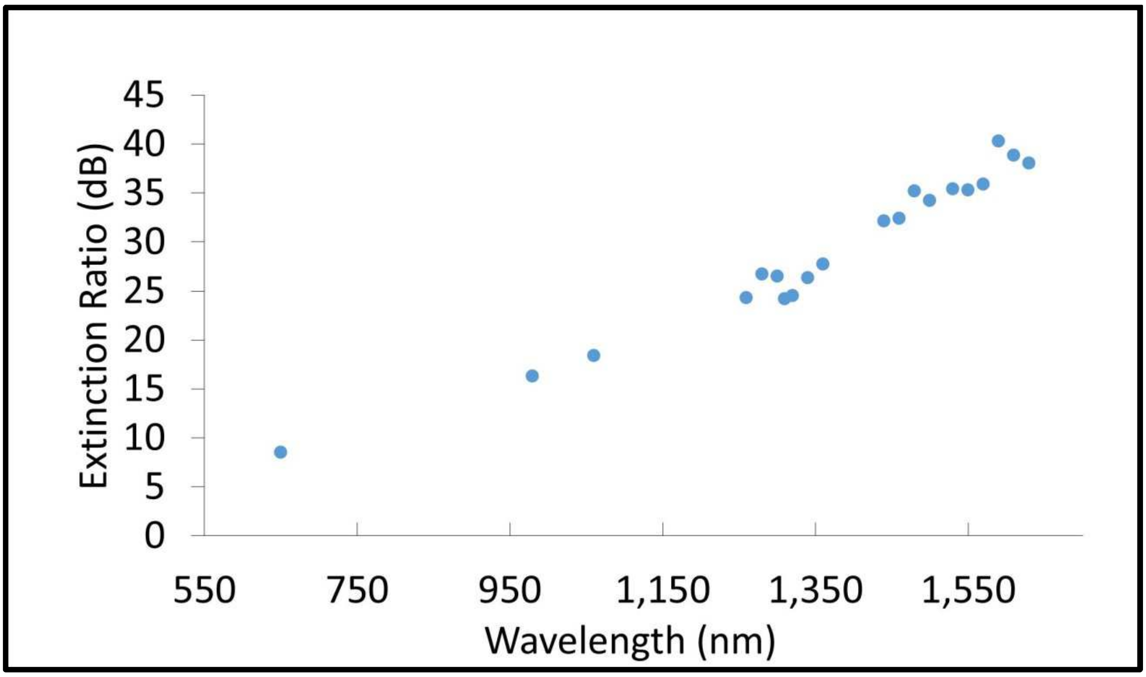

Figure 6.11: Broadband response of GO waveguide polarizer, where an extinction ratio of more than 20 dB is measured from 1250 – 1640 nm.

6.6 Simulation and Modelling

To verify and confirm the experimental observations and results, the field distributions of the orthogonally polarized modes are simulated using Finite Element Method (FEM) computation based on information from the SEM images and from the literature. Figure 6.12 illustrates the cross-section of the GO waveguide polarizer used in the simulation, with air gaps between the GO film and waveguide channel sidewall being taken into consideration. The typical waveguide cross-sectional dimensions used are a rectangular cross section 5 μm high and 10 μm wide. The GO film thickness is varied in direct relation to the number of GO solution drops being applied. An air gap with a maximum width at the base of 0.5 μm is inserted at each side of the waveguide channel sidewall. A simplifying assumption made in the simulation is that the GO film thickness is uniform across the entire coating region of interest.

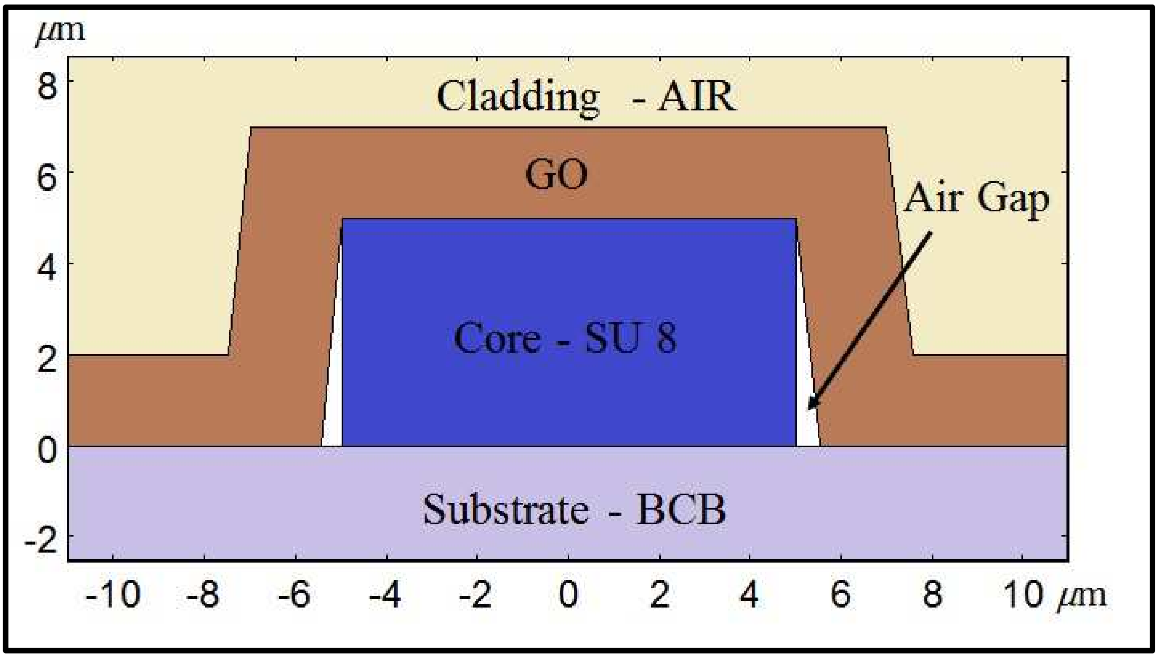

Figure 6.12: Illustration of GO-coated waveguide channel cross section. The waveguide channel cross section has a dimension of 5 μm in height and 10 μm in widths. GO coating thickness depends on the number of GO solution applied during drop-casting. The maximum air gap between the waveguide channel sidewall and the GO film is 0.5 μm at the base of the channel.

Figure 6.13 shows the modal distributions for both the TE- and TM-polarized modes, with the GO film thickness set at 2 μm. It can be seen that a fraction of the modal field was coupled from the waveguide channel into the GO film above the waveguide. Due to the highly anisotropic complex dielectric function of the GO coating, the orthogonal modes will 'see' the GO film very differently. A (quasi-) TM-polarized mode will see the GO film as a simple and relatively low-loss dielectric medium. Low loss propagation is therefore possible in this layer. On the other hand, a (quasi-) TE-polarized mode will see a GO film with a relatively large optical frequency conductivity value, giving strong damping effects and therefore high-loss propagation in the GO film. Note that, despite the existence of the wedge-shaped air gaps between the GO film and the waveguide channel sidewall, only a small portion of the light in the waveguide is coupled into the GO multi-layer across the air gaps, as shown in the modal distributions of both the TE- and TM-polarized modes in Figure 6.13 (bottom). In this case, the (quasi-) TM-polarized mode will experience high-loss propagation while the (quasi-) TE-polarized mode will see the GO film as a relatively low-loss dielectric medium.

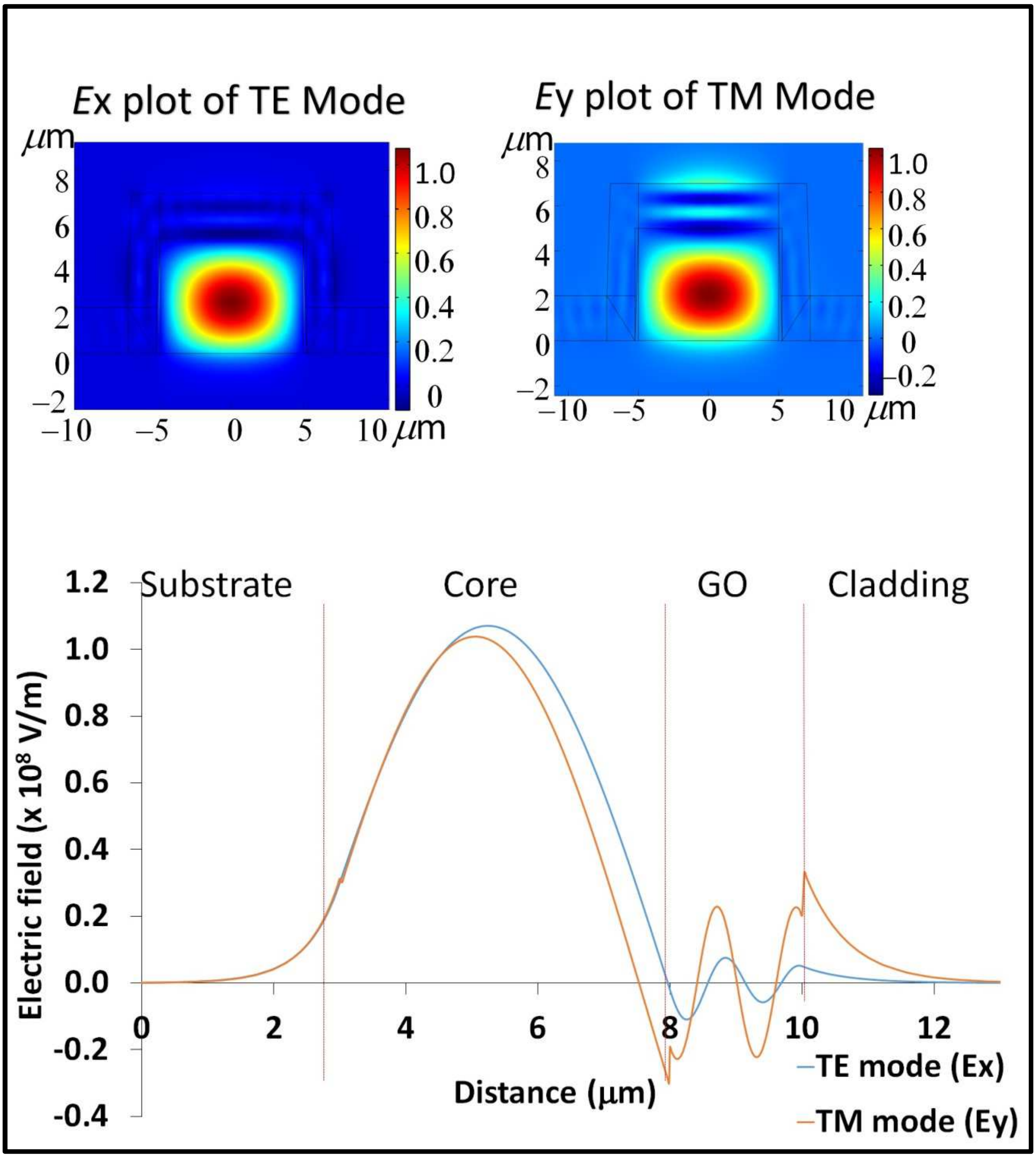

Figure 6.13 (Top & bottom): The simulated modal electric field component polarized field in the GO film, while the TE-polarized light coupled into the GO film is highly damped. The GO thickness used is 2.0 μm. The wavelength used in the simulation is 1550 nm.

For the simulation of the guided-light propagation in the GO-coated polymer waveguide, the choice of values for the real and imaginary parts of the anisotropic complex refractive index of the GO multi-layer is consistent with values published in the literature. For the component of the optical electric-field that is normal to the GO planes, the imaginary part of the refractive index may be neglected. This choice is justified by the fact that electron transport in this direction will be substantially blocked on the atomic scale by the barriers between nearest-neighbour graphene layers formed by layers of oxygen atoms as well as atoms of any other element that may be present. The real part of the refractive index has been set to 2, corresponding to a value of 4 for the real part of the dielectric function. This choice of n = 2 is in reasonable conformity with the values (~1.9) given by Vaupel and Stoberl [6] and is an intermediate value by comparison with refractive index values that would be obtained from the low-frequency (quasi-dc) values for the (relative) dielectric function that are given by Loh *et*. *al*. [7].

For the optical electric field component that is parallel to the GO layers and hence to the atomic sheets of graphene, the situation is substantially different. Because of the sub-wavelength scale of the individual GO layers, an effective (average) medium approach is appropriate. For the real part of the refractive index, the same value as for the perpendicular direction is chosen. The choice for the imaginary part reflects the fact that the graphene atomic sheets, even though they are fully (or almost fully) oxidised, are still capable of significant levels of conduction. Choice of an optical-frequency conductance value of σ = 2700 $S.m^{-1}$ implies a significant absorption coefficient for TE-polarized light. This value for the conductance is two orders of magnitude lower than that given by the usual Kubo expression for pure graphene (~1.8×10^5 $S.m^{-1}$). It is also less than the value of 9.0×10^4 $S.m^{-1}$ that applies for THz frequencies in reduced graphene oxide (rGO) [8]. With these choices for the refractive indices, the effective refractive index of the guided mode is obtained via simulation. By assuming a uniform coating of GO film, the optical propagation loss of the TE- and TM-polarized modes can be calculated using the Beer-Lambert law. Close agreement between simulation and experiment is obtained, as shown in Figure 6.14. The specific choices for the complex refractive indices should be taken as being indicative, rather than absolute. They are physically reasonable and are sufficiently accurate for identification of the characteristic behaviour that is found in our experimental measurements.

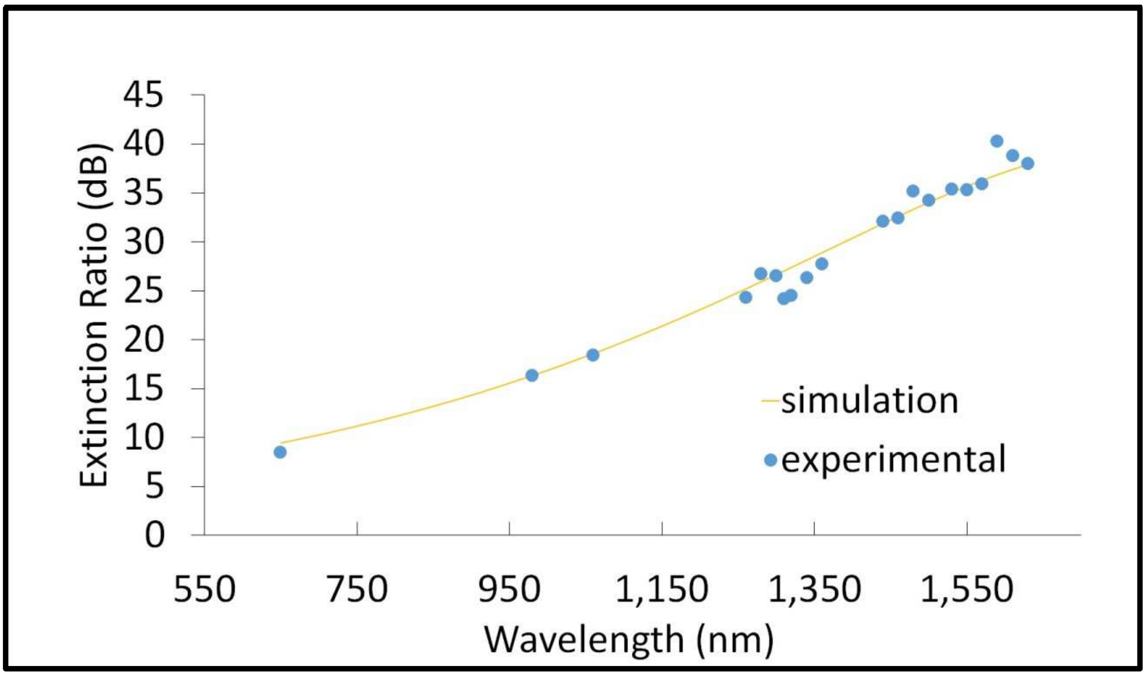

Figure 6.14: Broadband response of GO waveguide polarizer, where an extinction ratio of more than 20 dB is measured from 1250 – 1640 nm. Close agreement between simulation and experiment is obtained.

The ability to introduce GO coating using the drop-casting method provides a simple and effective means for waveguide polarizer fabrication. The performance of the GO waveguide polarizer can be improved using the coat-and-etch method demonstrated by Kim *et*. *al*. [4], or (for example) a laser ablated waveguide [9], where the GO film will only be coated on top of the channel waveguide. In addition, this method also enables the flexibility of selective areas drop-casting on an optical circuit intended for the polarization function, without the need for a physical mask during deposition or mechanical transfer of the graphene layers. With the use of an automated micropipette positioning and dispensing process, accurately localized deposition of GO coatings could be achieved in volume production.

6.7 Summary

A broadband waveguide polarizer with high extinction ratio has been demonstrated in a polymer waveguide coated with GO film deposited using the drop-casting method. Drop-casting of GO solution results in a substantially ordered GO layer stack, with its thickness increasing as the number of GO droplets applied increases. The polarization effect of the GO waveguide polarizer has been shown to be a result of the anisotropic complex dielectric function of GO film. The extinction ratio of the GO polarizer is dependent on the GO film thickness. The average extinction ratio is 38 dB between 1530 nm and 1630 nm, with the highest extinction ratio of 40 dB measured at 1590 nm and the lowest value of 35.5 dB measured at 1530 nm. The extinction ratio is achieved with only ~1.3 mm of GO coating length along the propagation direction. The short interaction length required to produce a high extinction ratio over a broad fibre-telecom wavelength band will provide a solution for the integrated waveguide polarizers required in applications that include optical lab-on-chip and photonic integrated circuits.

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