OPTIMIZATION OF THE GRAPHENE-SILVER BASED SURFACE PLASMON RESONANCE (SPR) SENSOR

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In this study, we investigated optical performance of a SPR-Sensor based on graphene-Silver substrate. Using the angle interrogation methods, we simulated the reflection spectrum which depends on the metal thickness and the refractive index of prism by exploiting the unique optical properties of graphene layers (especially in terms of its good absorption of molecules and its oxidation resistance). The addition of a certain number of graphene layers based on the knowledge that silver oxidation decreases the sensitivity of the sensor improved the sensitivity, while the detection accuracy decreased. To optimize the sensor performance, the investigation focused on monolayer graphene. Furthermore, genetic algorithms were used to optimize the SPR biosensor reflection by adjusting the coefficients of the system, which are the incidence angle and metal layer thickness.

Keywords: Surface plasmon resonance (SPR); biosensor; Graphene; genetic algorithm

1. Introduction:

Since last few decades, the biosensors based on the surface plasmon resonance technique have secured unique place, specifically in the determination of biomolecular interactions [1, 2].

Surface plasmon is a charge-density oscillation propagating along the interface between metal and dielectric medium. The evanescent wave of the surface plasmon is affected and determined by the properties of the metal and dielectric, and it can be excited by light. The Kretschmann configuration is one of the methods that are used in this excitation [3]. It is based on the observation of the reflected light spectrum obtained either by “angular interrogation” or “wavelength interrogation”. Generally, in SPR sensors the angular approach is the preferred due to its low angular resolution [4,5,6]. It is based on the measurement of changes in the resonant angle extracted from the curve when changing the angle of incidence and keeping the wavelength constant. The spectre is characterized by other factors: half width at half maximum (FWHM) and reflection minimum $R_{\min}$ [5].

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The evanescent waves propagating along the interface are extremely sensitive to several design parameters [7] as the presence of noble metals [8]. Ag is important because it supports the propagation of surface plasmon polariton waves (SPP) at visible light wavelengths [4,9]. It seems to be the most appealing for several reasons. It is not as expensive as Indium (*In*), nor reactive as sodium (*Na*) [10]. It provides in the metal-sensing layer interface a high value of electric field enhancement with low imaginary part of refractive index [11]. When compared with gold, the plasmon coupling in the silver has a sharper angular resonance peak [12], but it presents a drawback that it can be easily oxidized, thus affecting the performance of SPR-sensors [11,12].

To exploit the high enhancement characteristics of silver and deal with its oxidation problem, graphene [13,14] has been proposed as an addition of layers to protect silver from oxidation [11,15], and hence preventing the oxygen from interacting with them by a strong structure ring [15,16]. Another positive point for a graphene layer is the capacity of efficient adsorption of biomolecules [17]. In this paper we shall present an ultra-stable high performance SPR sensor based on a graphene on Ag configuration, which depends on the thickness of the silver layer, refractive index of prism and the number of graphene layers.

In the light of new optimization techniques, we used a genetic algorithm (GA), which is a heuristic method. It was found to be a powerful tool to solve and optimize problems based on the survival of the fittest. The theory of the evolution of species in their natural environment is an artificial transposition of basic concepts of genetics and survival of laws set forth by Charles Darwin: the most adapted individuals survive and reproduce. These same mechanisms will be used in the implementation of the GA [18,19], which will be validated by a problem which searches for the optimum reflectivity of graphene-SPR biosensor. The optimization procedure starts with the acquisition of different data related to the parameters in the problem, which are mainly the interval of incidence angle and the metal thickness layer. A good agreement is observed between the theoretical generalized multi-layer model and the optimization result. The obtained results allow us to conclude that the reflection fitness function is minimal whatever the number of iterations.

2. Theory

The SPR sensor with the Kretschmann configuration consists of a glass prism, a thin metal film, and a sample in sequence [20]. It is a simple model for excitation SPR, that understood with observed the SPR curve (the change reflected intensity R versus function angle incidence). When the light wave traverses the prism it generates an evanescent wave that propagates and penetrates in the metal surface with propagation constant k_x as in (1).

$$k_x = \left(\frac{2\pi}{\lambda_0}\right) n_1 \sin\theta \tag{1}$$

Where θ is the angle of incidence of light with the normal to the interface, and n_1 is the prism refractive index.

For a specific angle of incidence, k_x can be adjusted to match the SPP and its interaction with the light wave influences the reflectivity R [21]. In the simulation of the SPR biosensor with a graphene-on-silver substrate (shown in Fig.1(a)), we used the same previous model to which a new element was added: the graphene layer is sandwiched between the silver and a sensing film layer, to absorb the biomolecules present in the water.

The composition of the sensor is as follows:

- A tunable laser (632.8 nm).

-A prism made of silica glass *BK7* (n_1=1.515, depth $d = \infty$).

- A silver metal layer whose dielectric function follows the Lorentz-Drude model. The relationship is given by.

$$\varepsilon_m(\omega) = 1 - \left(\frac{f_0\omega_p^2}{\omega(\omega + i\Gamma_0)}\right) + \sum_{j=1}^{k} \frac{f_j\omega_p^2}{\omega_j^2 - \omega^2 + i\omega_j} \tag{2}$$

Where $\omega = \frac{2\pi c}{\lambda}$ is the light frequency, ω_p the plasma frequency, k the number of oscillators with frequency ω_j, strength f_j, and lifetime $\frac{1}{\Gamma_j}$; while $f_0\omega_p^2$ is the plasma frequency associated with intra-band transitions with oscillator strength f_0 and damping constant Γ_0 [20].

- A graphene layer of refractive index $n_{gr} = 2.7 + i1.4$ [22] and thickness $d = L \times 0.34nm$ [23], where L is the number of graphene layers.

- A bio-sensing layer $(n_s = 1.33)$.

We will use a generalized N-layer model [1, 20], shown in Fig. 1(b). To generate a SPR curve and obtain the reflectivity of the incident p-polarized light [22], all the layers are stacked along the z-direction. The arbitrary medium layer is defined by the thickness (d_k), dielectric constant (ε_k), permeability (μ_k), and refractive index (n_k). The tangential fields at the first $z = z_1 = 0$ and at the last boundary $z = z_{N-1}$ [1] are related by:

$$\begin{bmatrix} U_1 \\ V_1 \end{bmatrix} = M \begin{bmatrix} U_{N-1} \\ V_{N-1} \end{bmatrix} \tag{3}$$

U_1, U_{N-1} are the components of electric field at the boundary of the first and Nth layers of structure, respectively, while V_1, V_{N-1} are the components of magnetic field and M is the characteristic matrix of this structure, which is given by.

$$M = \prod_{K=2}^{N-1} M_K \tag{4}$$

With

$$M_k = \begin{bmatrix} \cos\beta_k & -i\sin\beta_k / q_k \\ -iq_k \sin\beta_k & \cos\beta_k \end{bmatrix} \tag{5}$$

Where

$$q_k = \frac{\left(\varepsilon_k + n_1^2 \sin^2\theta\right)^{1/2}}{\varepsilon_k}$$

$$\beta_k = \frac{2\pi d_k}{\lambda}\left(\varepsilon_k + n_1^2 \sin^2\theta\right)^{1/2} \tag{6}$$

The amplitude reflection coefficient (r_p) for p-polarized light is:

$$r_p = \frac{(M_{11} + M_{12}q_N)q_1 - (M_{21} + M_{22}q_N)q_N}{(M_{11} + M_{12}q_N)q_1 + (M_{21} + M_{22}q_N)q_N} \tag{7}$$

The reflection coefficient of the N-layer model for p-polarized light is:

$$R_p = \left|r_p\right|^2 \tag{8}$$

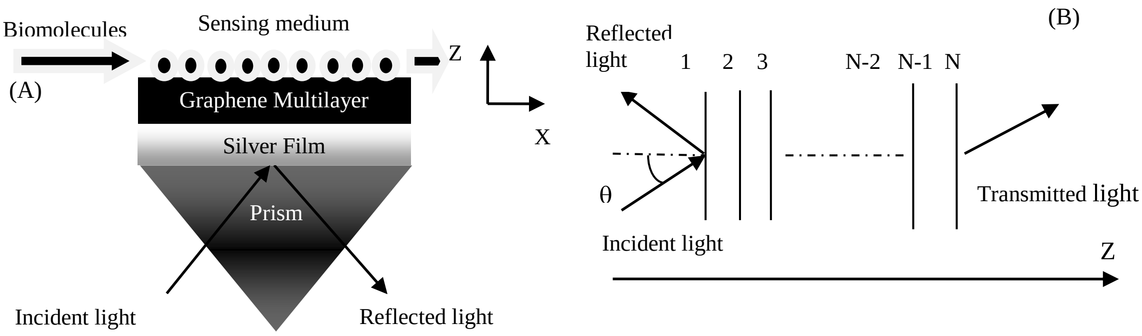


Fig. 1 (a) Configuration of the graphene-on-silver SPR biosensor [1]. (b) Schematic diagram of the multi-layer model [20].

For an enhanced performance of biosensors, the sensitivity is an important issue for its application. It depends on a number of factors such as the operating wavelength and material characteristics, including the refractive index of dielectric layer, refractive index of prism, metal film, and film thickness, etc. They are selected to optimize the resonance condition [2]. The sensitivity of the sensor is defined as [1]:

$$S = \left(\frac{\Delta \theta_{res}}{\Delta n} \right) \tag{9}$$

where the change in the resonance angle is $\Delta\theta_{res}$, corresponding to a change Δn in the refractive index of sensing layer. The resonance angle (θ_{res}) corresponding to the minimum reflection coefficient $(R_{\min})$ is determined from the $R-\theta$ curve.

Another important requirement for a high-performance SPR sensor is its high detection accuracy (which is inversely proportional to the FWHM). It can be noted that the precision of the measured SPR angle depends strongly on the width of the SPR curve [11,24].

3. Design parameters and results

3.1 influence of the metallic layer thickness on SPR response

The metal thickness is a very important parameter in surface Plasmon resonance. It defines the maximum dynamic and shape of the Plasmon [7,20].

In order to study the effect of the film thickness on the SPR-graphene sensor, we should take into account the number of graphene layers. Fig. 2(a) illustrates that when the number of graphene layers L increases, it leads to a shift of the resonance peak, a variation of its amplitude, and a change and modification of the width of its peaks which become wider respectively. For five graphene layers, the plasmonic angles shift from 67.81º to 69.02º. Therefore, it was shown in Fig. 2(b) that the change of the position of the minimum surface plasmon varies linearly with the number of additional graphene layers.

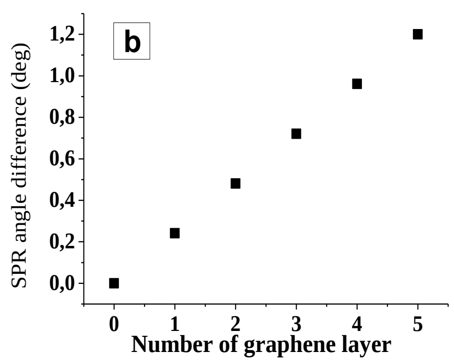


Fig. 2(a) SPR reflectance and (b) change in the SPR angle as the number of graphene layers increases (0-5 layers), with sensing layer refractive index of 1.330

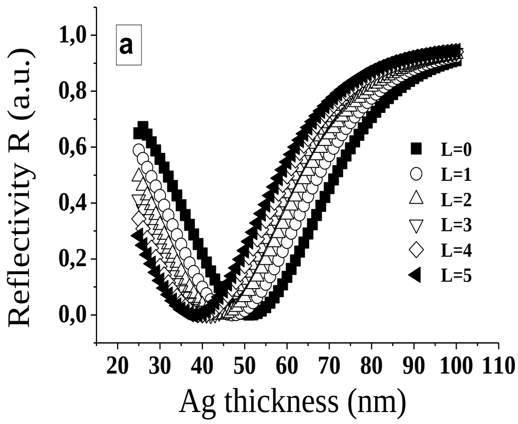
Additionally, one can note that the reduction in the amplitude of the resonance dip is a direct consequence of the change in the propagation constant of surface plasmons as a function of the number of graphene layers. The latter becomes different of the wave vector tangential component of incident light and hence the resonance condition is not fulfilled (i.e., SPs become damped) [10,17]. To minimize this effect, we optimize the silver layer thickness that decreases when the number of graphene layers increases (see Table 1).

Table 1.

Optimized values of silver thickness in terms of the number of graphene layers for corresponding changes in the resonance angle for zero reflectivity in the SPR. The refractive index of the sensing film was 1.33.

L GRAPHENE LAYERS NUMBER	D_{Th} SILVER THICKNESS (10^{-9}NM) THEORY	$\theta_{Th}\,(SPR)$ THEORETICAL RESONANCE ANGLE(DEGREE)	$R_{\min(Th)}$ MINIMUM REFLECTION COEFFICIENT (A.U.)
0	51.26	67.817	6.4485E- 004
1	47.22	68.058	6.3873E-004
2	44.23	68.298	4.2867E-004
3	41.84	68.538	2.0990E-004
4	39.85	68.778	5.8913E-005
5	38.14	69.099	5.5688E-007

The silver thickness for which the reflectivity is zero represents the optimum thickness. Fig. 3(a) illustrates the variation of reflectivity as a function of silver layer thickness for various numbers of graphene layers.



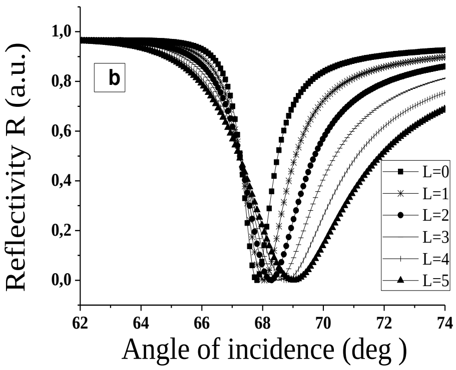


Fig. 3(a)Variation of reflectivity with respect to the thickness of the silver layer. (b)Angular reflectivity spectra corresponding to optimum values of the thickness of silver for graphene layers (1–5 layers)

Fig. 3(b) shows the angular spectra of reflectivity corresponding to these optimum values. It is noted that the optimization of the silver thickness provides a shallower dip in the reflectivity curve. The results are summarized in Table 1.

3.2. Graphene layers effect on detection accuracy and SPR-sensor sensitivity enhancement

It is important to make a good choice for the maximum number of graphene layers, which must be fixed for the high performance of the biosensor. This fixed number must be taken in order to give a high sensitivity enhancement and a high detection accuracy.

In Fig. 4(a), the sensitivity enhancement as a function of the number of graphene layers *L* after the absorption of biomolecules is plotted, assuming the same refractive index change Δn = 0.005. It is found that adding more graphene layers allows an increase in sensitivity. The sensitivity enhancement value can be 1.4% for the monolayer graphene biosensor, 3.1% (L=2), 4.5% (L=3), 6.1% (L=4), and 9.3% (L=5). Graphene is a good absorbent of biomolecules, since the increase of graphene layers means the increase in the absorption of biomolecules, thus an increase in sensitivity [10]. Fig. 4(b) shows the effect of the number of graphene layers on detection accuracy. We can see that the increase in the number (or thickness) of graphene layers causes a reduction in detection accuracy from 0.818 to 0.221degree^{-1}.The reason of this decrease is the breadth in the curve, as a result of the damped extension of SP field into the graphene layer, due to the amortization in the absorption of surface plasmon at the silver/graphene interface, which is also due to the optical properties of graphene (non-zero imaginary refractive index part) [10,17,21]. Finally, it is clear that the use of graphene as a protective layer in the Kretschmann configuration based on silver becomes important if the number of layers decreases.

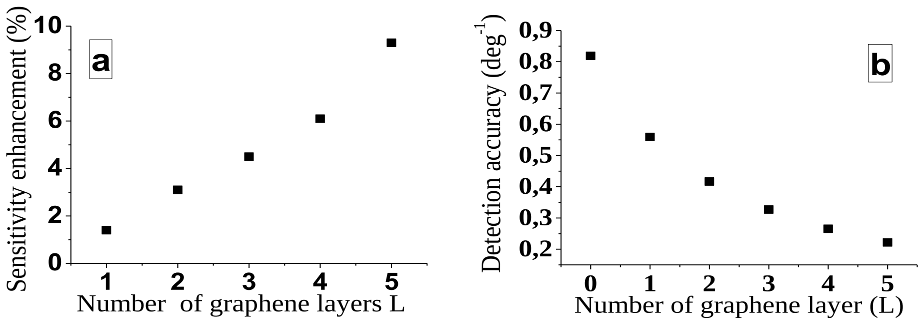


Fig. 4(a) Variation in sensibility enhancement and (b) detection accuracy versus the number of graphene layers (1-5 layers).

For the monolayer graphene SPR-sensor, we have theoretically demonstrated that the refractive index of the sensing film has an important influence on the reflection spectra and detection accuracy, because any change in the properties of the dielectric medium are determined by the surface plasmon evanescent waves [5]. Fig. 5 shows that when the sample refractive index n_s changes by 0.04 RIU (from 1.33 to 1.37), the width of resonance values increases from 1.7881 to 2.3642. This increase in the width of the curve leads to difficulties in a θ_{SPR} measurement [10,21], and therefore to a decrease in the accuracy detection of the refractive index measurement, as depicted in Fig.6. The results are compared with those obtained by P. K. Maharana *et al.* (2013) [22] for an operating wavelength of 633 nm. As it can be seen, they are in good agreement with the same change of sensing layer refractive index.

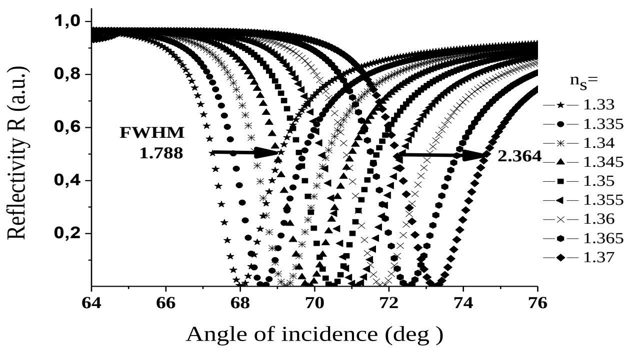


Fig. 5. SPR reflection spectra for monolayer graphene layers as a function of the incident angle with various values for sensing medium refractive index.

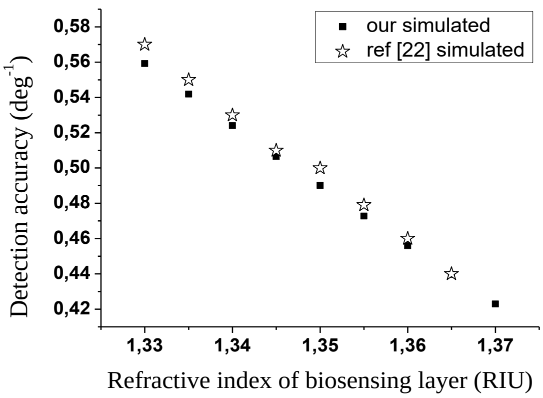


Fig. 6. Variation of detection accuracy with sensing layer refractive index for monolayer graphene at two wavelengths (632.8 nm in our study and 633 nm in Ref. [22]) for comparison.

The previous results appeared that the optimization of the silver layer thickness gives very deep peaks of resonance, but it cannot improve the detection accuracy. To overcome this problem, a high refractive index prism as chalcogenide prism (n_{2S2G} = 2.358 at λ = 632.8) [10, 25]. Fig.7 (a) show the angular reflectivity spectra corresponding to the optimal values of silver thickness in the SPR-graphene biosensor based on the chalcogenide prism, we can see that the high refractive index of the 2S2G glass enhances the deep and the width of the reflectivity curves. Also, compared with SPR-graphene biosensor based BK7 we can observed that when the graphene layers increases up to five layers, the resonance linewidth increses from 0.48°(1.2°) to 2.16°(4.5°) as illustrated in fig.7(b), This indicates that SPR-Biosensor based chalcogenide prism has better accuracy and then better resolution.

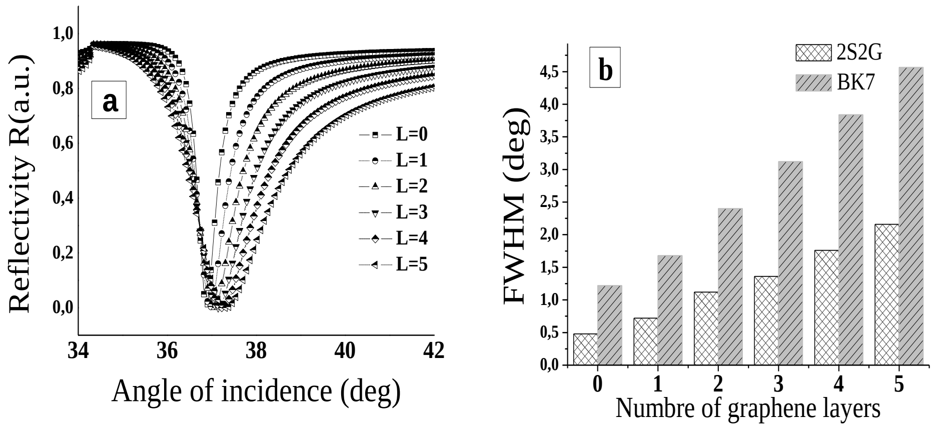


Fig. 7 (a) Angular reflectivity spectra corresponding to the optimal values of silver thickness in the SPR-graphene biosensor based on the chalcogenide prism (b). Resonance linewidth as a function of graphene layers number for BK7 and 2S2G prism.

4. Optimization of the Resonance angle and of reflection coefficient, and of the Silver thickness by using a Genetic numerical algorithm

The GAs are searching processes based on the laws of natural selection and genetics, from Darwinian's theory of survival of the fittest. They use probabilistic transition rules instead of deterministic rules. The GAs are iterative stochastic algorithms that work upon groups of codified points over an initial population. The population of potential solutions is represented by individuals or chromosomes (parents) that evolve through successive iterations called generations. The fittest individual will survive a generation and generate offspring, which is evaluated by the quality of its fitness function and with a similar fashion to their parents. This is the last individual, which might be stronger. In order to allow a population of solutions to converge to optimal solutions, we follow iteration process steps based on the genetic triangle: reproduction, evaluation, and selection [26-29]. In this work, we will be searching for the best value of resonance angle and thickness film, which will give the optimum reflection fitness function.

4.1. Synthesis and analysis of results

The development of the optimization procedure imposes on one side the development of a procedure to solve the characteristic matrix for an N-layer system (3), and on the other side, an implementation of the GA procedure. In this work, we intend to optimize the SPR-graphene sensor from the beginning, at the acquisition of different data related to the parameters denoted in the problem, which are mainly: wavelength λ (632.8nm) | prism (1.515)| Ag film layer (ε_{Ag} = -17.81+i0.676) | graphene ($d_{gr}=L$*0.34nm, $n_{gr}=2.7+i1.4$)|water. *L* is the number of graphene layers and is equal to one because, as we have seen previously, it is appropriate for the best performance of our sensor. The intervals of incidence angle θ_{inc} and thickness metal layer D are [10,90] (degree) and [30,100] (nm), respectively. The obtained results considering these parameters are summarized in Table 2.

Table 2.

Summary of results of the GA application

Population size	20
Number of generations	51
Method used in selection	Roulette wheel scheme (RWS)
Crossover probability P_C	0.8
Mutation probability p_m	0.01
Optimum minimum reflection coefficient R_{min} (a.u.)	4.3591e-10
Resonance angle optimize $d\theta_{Ag}$ (SPR)(degree)	67.927
Silver thickness optimized D_{Ag} (*10^{-9} nm)	48.404
Computing time (s)	≈0.732634

The optimized results of the problem clearly reveal the existence of a relation between the yielded results and the parameters of the GA (size of population, number of generations, probability of crossing, and probability of mutation).

The convergence of the GA for optimum reflectivity is represented by the best parameters found in Table 2. The choice of a population with a small size leads to a fast resolution, even with a large number of evaluations of the fitness function, and it is effective in terms of time. Therefore, no change in the metal thickness or resonance angle after these values of size was noticeable.

From Fig. 8, it can be seen that, independently on the number of iterations, the obtained results allow us to conclude that the reflection fitness function is always minimal and close to zero. Here, the optimum value with a weak error rate is determined by running the algorithm for 51 times. These results are achieved by optimizing both the values of the thickness of silver layer and incidence angle. One can note that there is an accomplishment in the rapprochement between the theoretical and optimized values.

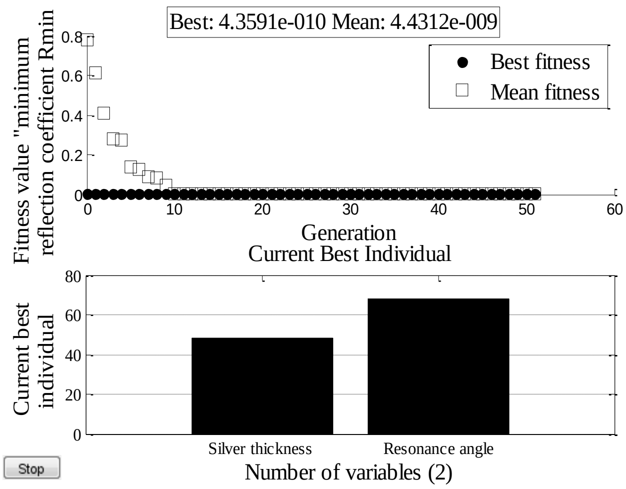


Fig. 8 Best fitness plot for the minimization of the reflection function. The plots show the results from Table2.

The GAs have the advantage to become better during its execution; the best potential solution is identified at the end of every generation and adopted with a certain strategy. This one consists of saving each time the best solution between the current and preceding one; in other words, it preserves only the best solutions [19].

5. Conclusion

We have investigated in this paper, the optical performance of a biosensor based on the phenomenon of surface Plasmon resonance (SPR) and including a layer of graphene sheet on the top of the silver layer. Through numerical simulations, we successfully demonstrated the significant roles of the silver layer thickness and prism refractive index in the presence of graphene layers which contribute to the remarkable shape of reflectivity curve, where the optimization of the silver layer thickness gives a very deep peaks of resonance and the high refractive index prism leads to deep and narrow resonance peaks. On the other hand, we have presented a discussion concerning the enhanced performance of SPR graphene sensor, which is assessed in terms of sensitivity and detection accuracy. We have found that the sensitivity enhancement value can be increased from 1.4% for the monolayer graphene to 9.3% for five layers, but the detection accuracy decreased from 0.78 to 0.46 degree^{-1}. Afterwards, we focused on the case of a monolayer graphene on a thin silver film used in the implementation of the GA. Its application has allowed us to obtain the searched results related to the problem of determination of the optimum reflected by optimal reflection, as well as seeking the best parameters allowing us to determine them according to the incidence angle and metal thickness.

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