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Graphene-MoS₂ with TiO₂—SiO₂ layers based surface plasmon resonance biosensor: Numerical development for formalin detection



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ABSTRACT

In this article, numerically a surface plasmon resonance (SPR) biosensor is developed based on Graphene- M_0S_2 with TiO2-SiO2 hybrid structure for the detection of formalin. Based on attenuated total reflection (ATR) method, we used angular interrogation technique to sense the presence the formalin by observing the change of "minimum reflectance with respect to SPR angle" and "maximum transmittance with respect to surface plasmon resonance frequency (SPRF)". Here, we used Chitosan as probe analyte to perform chemical reaction with formalin (formaldehyde) which is consider as target analyte. Simulation results show a negligible variation of SPRF and SPR angle for improper sensing of formalin that confirms absence of formalin whereas for proper sensing is considerably countable that confirms the presence of formalin. Thereafter, a comparison of sensitivity for different sensor structure is made. It is observed that the sensitivity without TiO2, SiO2, MoS2 and Graphene (conventional structure) is very poor and 73.67% whereas the sensitivity with graphene but without TiO2, SiO2 and MoS₂ layers is 74.67% consistently better than the conventional structure. This is due to the electron loss of graphene, which is accompanying with the imaginary dielectric constant. Furthermore, the sensitivity without TiO2, SiO2 and graphene but with MoS2 layer is 79.167%. After more if both graphene and MoS2 are used and TiO₂ and SiO₂ layers are not used then sensitivity improves to 80.5%. This greater than before performance is due to the absorption ability and optical characteristics of graphene biomolecules and high fluorescence quenching ability of MoS₂. Further again, if TiO₂-SiO₂ composite layer is used with the Graphene-MoS2 then the sensitivity enhances from 80.5% to 82.5%. Finally, the sensitivity for the proposed structure has been carried out, and result is 82.83%, the highest value among all the previous structures to integrate the advantages of graphene, MoS2, TiO2 and SiO2.

1. Introduction

Formalin (40% formaldehyde) is a toxic element soluble in water, has been categorized as *Group I* Carcinogen to human beings by the International Agency for Research on Cancer (IRAC) [1]. Recent news has claimed the use of formaldehyde in food preservation that is very popular, particularly in Asian countries [1]. As a result, the finding of formalin is a worried issue which is a biochemical process. Its mechanism of action for fixing lies in its ability to form cross-links between soluble and structural proteins. The resulting structure retains its cellular constituents in them in vivo relationships to each other, giving it a degree of mechanical strength which enables it to withstand subsequent processing, as reported by Environmental and Occupational health and

Safely Services 2004 [2].

Today, Biosensors have been penetratingly researched owing to their importance of many industry applications such as medical diagnosis, enzyme detection, food safety and environmental monitoring [3,4]. Today numeral biosensors have been scientifically advanced, among them surface plasmon resonance (SPR) biosensor accepts the advantage of compactness, light weight, high sensitivity, the case of multiplexing and remote sensing and so forth [5]. The SPR sensor works on the basis of attenuated total reflection (ATR) method basically in angular interrogation technique. The ATR method practices a property of total internal reflection (TIR) resulting in a momentary wave normally known as surface plasmon waves (SPW). A beam of incident light is passed through the ATR crystal in such a way that it reflects at least

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once off the internal surface in contact with the sample. This reflection forms the momentary wave which extends into the sample [3]. SPW is a momentary guided electromagnetic wave that propagates along a metal-dielectric interface by utilizing the surface plasmon waves (SPW). The variation of the biomolecules concentration on account of chemical reaction, will produce the local modification of the surrounding refractive index (RI) near the sensor surface that outcomes in altering the propagation constant of the SPW and thus the SPR angle and SPR frequency (SPRF) changes [6]. The SPR technique has been successfully applied in various fields, such as chemical and biochemical sensing, film characterization and beam characterization.

Many conventional methods are available for the detection of formalin such as Gas chromatography-mass spectrometry (GC-MS), high performance liquid chromatography (HPLC), fluorimetry, Nash test, gravimetric methods and other chemical based biosensors [7,8,9,10]. Colorimetric detection methods such as Deniges and Eegriwes methods have been known since the beginning of the 20th century [6]. Unluckily, these methods, reagents and reaction products are often just as harmful to human health. All of these conventional methods require similarly hazardous reagents and suffer from a number of interferences, resulting in false positions. Additionally, these methods are impracticable for real time measurements [1].

In this article, numerically Graphene-MoS2 with TiO2-SiO2 layer based SPR biosensor is designed for formalin detection by observing the change of SPR angle-minimum reflectance and SPR frequency-maximum transmittance. Here, graphene and MoS2 are used as biomolecular acknowledgement analytes (BAA), TiO2-SiO2 bilayer as the improvement of sensitivity and Gold (Au) as the sharp SPR curve and the oscillation of surface electrons. The enlargement of SPR angle and frequency reasons an increase of SPR performance (sensitivity) [12]. This sensor is identified the presence the formalin based on molecular concentration that is varied due to the immobilization of chitosan (probe analyte) on the sensor surface that changes the RI of sensing analytes. The RI modification will in turn prime to modify in the SPR angle and SPR frequency that explains a final change in propagation constant of SPW [6]. Finally, it is shown that the sensitivity of conventional SPR sensor is 73.67% and the graphene-MoS2-based sensor is enhanced to 79.167%. with respect conventional SPR sensor. The sensitivity is further enhanced to 82.83%, by including TiO2-SiO2 composite layer with respect to conventional SPR sensor. At the end of this letter, a comparative study of the sensitivity of the proposed work with the existing works is discussed.

2. Methodology

A schematic diagram of the proposed composite layered SPR biosensor is shown in Fig. 1. On the basis of kretschmann configuration of SPR, composite layer of Graphene-MoS2-Au-TiO2-SiO2 have deposited on the base of prism and this whole arrangement kept in contact with the PBS or sample containing the target biomolecule/chemical also known as analytes, for sensing application [12]. We used Fresnel seven layered optical system to model the proposed sensor which is elaborately discussed in literature [6,11]. By defining the sensor layers, the first layer is SF11 glass prism (RI, $n_p = 1.7786$) [12], second layer is TiO_2 (RI, $n_2 = 2.5837$) [13], third] layer is SiO_2 (RI, $n_3 = 1.4570$) [12], fourth layer is Au (RI, $n_4 = 0.1838 + i*3.4313$) [14], fifth layer is MoS_2 (RI, $n_5 = 5.9 + i*0.8$) [12], sixth layer is graphene (RI, $n_6 = 3.0 + i 1.1487$ [12] and final layer is Phosphate buffer saline (PBS) solution(RI $n_7 = 1.34$) as bare sensing dielectric medium that's affords better adsorption of biomolecules [11-21]. After completing the modeling, a TM polarized He-Ne (wavelength = 633 nm) light wave (optical wave) is used to incident as shown in Fig. 1, which passes through the prism and some portion is reflected at the prism-gold interface. During intruding light energy to prism-gold interface, a momentary wave is generated which is known as surface plasmon wave (SPW) mentioning in introduction section. This SPW is reflected at the

prism-gold interface. The reflection intensity for TM-polarized light is expressed as [11]:

$$R_p = |r_p|^2 \tag{1}$$

Where,
$$r_p = \frac{(F_{11} + F_{12}n_N)n_1 - (F_{21} + F_{22}n_N)}{(F_{11} + F_{12}n_N)n_1 + (F_{21} + F_{22}n_N)}$$
 (2)

In equation (2), F is defined as [11].

$$F_{ij} = \left[\prod_{k=2}^{4-1} \begin{pmatrix} \cos \beta_k & -\frac{i \sin \beta_k}{n_k} \\ -i n_k \sin \beta_k & \cos \beta_k \end{pmatrix} \right]_{ij} = \left[F_{11} & F_{12} \\ F_{21} & F_{22} \right]$$
(3)

Here n_k is arbitrary transverse refractive indices of the corresponding kth layer which can be explained by the relation [11]:

$$n_k = \left[\frac{\mu_k}{\varepsilon_k}\right]^{1/2} \cos \theta_k = \sqrt{\frac{\varepsilon_k - (n_p \sin \theta_{in})^2}{\varepsilon_k^2}}$$
(4)

And β_k is arbitrary phase constant of the corresponding kth layer which can be explained by the relation [11]:

$$\beta_k = \frac{2\pi}{\lambda} n_k \cos \theta_k (z_k - z_{k-1}) = \frac{2\pi}{\lambda} d_k \sqrt{\varepsilon_k - (n_p \sin \theta_{in})^2}$$
 (5)

Here, z_k is the wave impedance of kth layer and is defined as [6]:

$$z_k = \frac{k_{light} n_k \cos \theta_k}{(2\pi c/\lambda_{633})\varepsilon_k^2} \tag{6}$$

And θ_k is the incident angle of kth layer, is defined as [6]:

$$\theta_k = a\cos(\sqrt{1 - (n_{k-1}/n_k)\sin^2\theta_{in}}) \tag{7}$$

In equations (3)–(7), n_p is the RI of prism, θ_{in} is the initial incident angle indicated in equation (1), ϵ_k is the permittivity of kth layer dielectric, d_k is the thickness of kth layer (thickness of graphene $d_g=L\times 0.34\,\text{nm}$, where L is the number of graphene layers, thickness of gold $d_{Au}=50\,\text{nm}$) respectively.

The SPW propagates with the dissimilar propagation constant from optical wave which is defined by Eq. (10). The propagation constant of SPW is varied due to the immobilization of formalin (target legend) into chitosan (probe legend which is presence in sensing analytes), and once being equal to the propagation constant of incident light. The point at which incident light propagation constant equals the SPW propagation constant is called SPR point [6]. Eq. (8) depicts that SPR angle is a RI dependent parameter of sensing medium. At SPR point, the frequency at which SPW propagates is called surface plasmon resonance frequency (SPRF) and the angle of incidence is called SPR angle that can be given as follows:

$$\theta_{SPR} = a \sin \sqrt{\frac{n_{com}^2 n_s^2}{n_p^2 (n_{com}^2 + n_s^2)}}$$
 (8)

Here, n_{com} refers equivalent RI of composite layer define as $n_{com} = \sqrt[5]{n_2 n_3 n_4 n_5 n_6}$ When formalin is flowing through chitosan on the sensor surface according to Fig. 1, then the RI of sensing medium is modified owing to performing chemical reaction as follows [6]:

$$n_s^2 = n_s^1 + C_a \frac{dn}{dc} \tag{9}$$

Here, n_s^1 is the refractive index (RI) of the sensing dielectric before adsorption of formaldehyde molecules. When no dielectric sample (probe or target) is present inside the sensing medium then n_s^1 is the RI of PBS saline ($n_7=1.34$) which is available in bare sensor. C_a is the concentration of adsorbed bio molecules, for example if 1000 nM concentrated formaldehyde molecules has been added into the sensing medium, then the value of $C_a=1000$ nM. The $\frac{dn}{dc}$ is the RI increment parameter, suppose, after adding 1000 nM concentrated formaldehyde molecules, the RI of sensing layer has been changed because the sensing layer now consists not only PBS but also formaldehyde. The changed

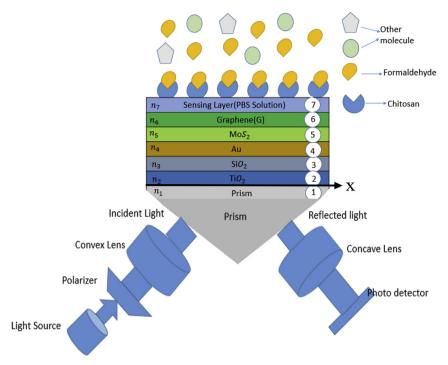


Fig. 1. Schematic of Graphene-MoS2-Au-TiO2-SiO2 model for mechanism of formalin detection with hybrid layer biosensor.

value of RI from PBS is $\frac{dn}{dc}$ (= 0.181 cm³/gm for PBS as bare case [11]). And n_s^2 is the RI of the sensing dielectric after adsorption of formaldehyde molecules.

If SPR angle changes, the propagation constant of SPW also changes which was explained mathematically in the literature [5] as given below:

$$K_{spw} = \frac{2\pi}{\lambda} n_p \sin \theta spr \tag{10}$$

and finally if propagation constant of SPW changes it makes the surface resonance frequency (SPRF) change which can be explained by the following equation:

$$SPRF = \frac{c_o}{n_{com}} \frac{k_{SPW}}{2\pi} \tag{11}$$

Where $\frac{c_0}{n_{com}}$ is the propagation velocity of SPW that is a perpendicularly confined evanescent electromagnetic wave [16,17]. If the incident angle of optical wave is tuned, SPR condition is achieved in which reflectance (R) of reflected wave is minimum and transmittance (T) is maximum and then SPW penetrate at SPF along the x-direction. We define two plot "transmittance versus surface resonance frequency (T ~ SRF curve)," as well as "Reflectance versus surface resonance angle (R ~ SPR-angle curve)," as surface resonance attributor.

3. Numerical results and discussions

Numerical analysis has been initiated to check the routine of proposed sensor by finding it surface plasmon resonance (SPR) angle versus the change of minimum reflectance (R \sim SPR-angle)" attributor and "the surface plasmon resonance frequency (SPRF) versus maximum transmittance (T \sim SPRF)" attributor curve. Fig. 2 (a) and (b), show R \sim SPR-angle and T \sim SPRF curve. The angle of incidence and SPRF of bare sensor are 56.26^{0} and 97.968 THz respectively. And the angle of incidence and SPRF while $1000\,\text{nM}$ probe chitosan are placed on sensing dielectric, are 56.34^{0} and 98.688 THz respectively. Results show no noteworthy change in SPR angle and SPRF (change of SPR angle is 0.08^{0} , change of SRF is 0.72 THz only) since there no bonding has taken place between probe and target due to the absence of formalin.

Fig. 3 demonstrates the concluding stage of detection concept. It shows, the change of attributor ($\theta sp \& R_{min}$) and ($\Delta SPRF \& T_{max}$) while 1000 nM formaldehyde molecule is sinking in the probe. Results suggest significant change in SPR angle as well as SPRF (58.05° and 99.875 THz) due to bonding has taken place between probe (chitosan) and target (formalin). So there is considerable change of charges in target molecule. The change of detecting attributor ($\theta sp \& R_{min}$) and ($\Delta SPRF \& T_{max}$) owing to adding formalin is provided in Table 1. The amount of shift rises with increasing concentration of the detectionable target from 1 to 200 nM as stated by Eq. (9) and tabulated in Table 1. The information of Table 1 has been extracted from Fig. 3(a) and (b).

Table 1 provides information about how θ SPR and SPRF parameters change with different concentrations of formalin molecules. It is apparently seen that the considerable increase of SPR angle and SPRF is a sign of bonding between probe and target. Upon making a bond with the target, the chemical configuration of legend is changed, which leads to the change in the optical properties. Hence thus we can observe whether is there formalin in the sample or not. Also increased amount of formalin forms more recurring bonds thus indicating greater interaction [19,20].

For finalizing a decision, at first we measure and tabulate the values of ΔR_{min}^{P-T} and $\Delta \theta_{sp}^{P-T}$ in Table 2 and compare these to threshold values $((\Delta R_{min}^{P-T})_{\min}$ and $(\Delta \theta_{sp}^{P-T})_{\min})$ If the measured values are greater than these threshold values, then we say there is presence of formalin in the sample. The following equations describe the threshold parameters:

$$(\Delta R_{\min}^{P-T})_{\min} = |R_{\min}^{probe} - R_{\min}^{T \operatorname{arg} et}| = 0.0018$$
 (12)

$$(\Delta \theta_{sp}^{P-T})_{\min} = |\theta_{sp}^{P-T} - \theta_{sp}^{T \arg et}| = 1.71$$
 (13)

where, $(\Delta R_{min}^{P-T})_{min}$ is the threshold value of minimum changed reflectance, $(\Delta \theta_{sp}^{P-T})_{min}$ is the threshold value of changed SPR angle, R_{min}^{Probe} represents the minimum reflectance of probe legend (chitosan), R_{min}^{Target} denotes the minimum reflectance of sampling target, θ_{sp}^{Probe} depicts the SPR angle of probe legend and finally SPR θ_{sp}^{Target} is the SPR angle of sampling target. We reached and tabulated the same calculation by taking $\Delta SPRF_{p-t}$ and ΔT_{max}^{p-t} as also the detecting attributors. The following equations are used to determine the threshold values of these attributors as:

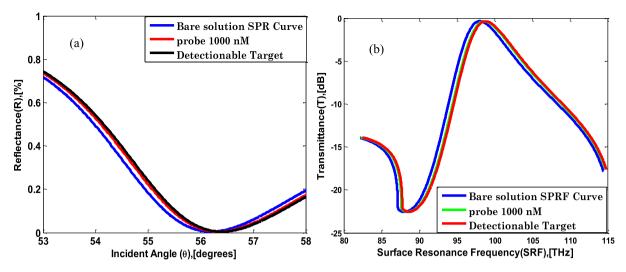


Fig. 2. Numerical results of Bare SPR Sensor (a) R ~ SPR-angle curve in the absence of formalin and chitosan. (b) T ~ SPRF curve in the absence of formalin and chitosan.

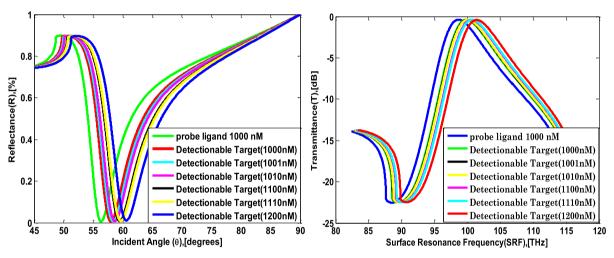


Fig. 3. (a) Reflectance vs. Incident Angle Curve and (b) Transmittance vs. SPR Frequency Curve for Different Concentration of Detectionable Target.

Table 1 $R_{min}[\%]$, $\theta_{SP}[\deg]$, $T_{max}[dB]$ and SPRF[THz] for different concentrated dielectrics medium.

Concentration (C _a) [nM]	R _{min} [%]	$\theta_{SP}[\deg]$	T _{max} [dB]	SPRF [THz]
1000 (immobilizer Probe) 1000 (Detectionable Target) 1001 (Detectionable Target) 1010 (Detectionable Target) 1100 (Detectionable Target) 1110 (Detectionable Target)	0.0044 0.0062 0.0066 0.0070 0.0082 0.0085	56.3400 58.0500 58.3800 58.6700 59.4900 59.6800	0.3795 0.3981 0.4002 0.4018 0.4106 0.4129	98.688 99.875 100.008 100.106 100.627 100.761
1200 (Detectionable Target)	0.0100	60.6200	0.4249	101.447

$$(\Delta T_{\text{max}}^{p-t})_{\text{min}} = |T_{\text{max}}^p - T_{\text{max}}^{t=1000nm}| = 0.0186$$
(14)

$$(\Delta SRF_{p-t}) = |SRFL_p - SRF_{t=1000nM}| = 1.187$$
(15)

The numerical data judges the strong dependency of the SPR angle and SPRF on the concentration increment that reflects in reflectance and transmittance characteristics curve.

These obtained numerical values can really give an idea about successful interaction or the failed ones. The first condition in Table 3 expresses the desired condition, second and third one needs careful recheck for attaining desired condition, fourth condition confirms the probe is still free from formalin molecule.

Table 2 Calculated ΔR_{min}^{P-T} [%], ΔT_{max}^{p-t} , ΔSRF_{p-t} [THz] and $\Delta \Theta_{sp}^{P-T}$ [deg] values from Eq. (5) to Eq. (8) for different concentration of dielectric medium.

Concentration (C _a) [nM]	$\Delta R_{min}^{P-T}[\%] = \left R_{min}^{Probe} - R_{min}^{Target} \right $	$\Delta\theta_{sp}^{P-T}[\text{deg}] = \theta_{sp}^{Probe} - \theta_{sp}^{Target} $	$\Delta T_{max}^{p-t}\left[dB\right] = \left. T_{max}^p - T_{max}^t \right $	$\Delta SRF_{p-t} [THz] = \left SRF_p - SRF_t \right $
1000 (Target)	$(\Delta R_{min}^{P-T})_{\min}$	$(\Delta heta_{Sp}^{P-T})_{\min}$	$(\Delta T_{ m max}^{p-t})_{ m min}$	$(\Delta SRF_{p-t})_{\min}$
1001 (Target)	0.0022	2.04	0.0207	1.32
1010 (Target)	0.0026	2.33	0.0223	1.418
1100 (Target)	0.0038	3.15	0.0311	1.939
1110 (Target)	0.0041	3.34	0.0334	2.073
1200 (Target)	0.0056	4.28	0.0354	2.759

Table 3Four probable conditions for making decision about successful interaction.

Conditions for using & $R_{\rm min}$ as detecting attributor	Conditions for using $\Delta SPRF\ \&\ T_{max}$ as detecting attributor	Decision
$\begin{array}{l} \Delta R_{min}^{P-T} \ \geq \ (\Delta R_{min}^{P-T})_{\min} \ \&\& \ \Delta \beta_{sp}^{P-T} \ \geq \ (\Delta \beta_{sp}^{P-T})_{\min} \\ \Delta R_{min}^{P-T} \ \geq \ (\Delta R_{min}^{P-T})_{\min} \ \&\& \ \Delta \beta_{sp}^{P-T} \ \leq \ (\Delta \beta_{sp}^{P-T})_{\min} \\ \Delta R_{min}^{P-T} \ \leq \ (\Delta R_{min}^{P-T})_{\min} \ \&\& \ \Delta \beta_{sp}^{P-T} \ \geq \ (\Delta \beta_{sp}^{P-T})_{\min} \\ \Delta R_{min}^{P-T} \ \leq \ (\Delta R_{min}^{P-T})_{\min} \ \&\& \ \Delta \beta_{sp}^{P-T} \ \leq \ (\Delta \beta_{sp}^{P-T})_{\min} \end{array}$	$\begin{array}{l} \Delta T_{max}^{p-t} \geq (\Delta T_{max}^{p-t})_{\min} \; \&\& \; \Delta SRF_{p-t} \geq (\Delta SRF_{p-t})_{\min} \\ \Delta T_{max}^{p-t} \geq (\Delta T_{max}^{p-t})_{\min} \&\& \; \Delta SRF_{p-t} \leq (\Delta SRF_{p-t})_{\min} \\ \Delta T_{max}^{p-t} \leq (\Delta T_{max}^{p-t})_{\min} \; \&\& \; \Delta SRF_{p-t} \geq (\Delta SRF_{p-t})_{\min} \\ \Delta T_{max}^{p-t} \leq (\Delta T_{max}^{p-t})_{\min} \&\& \; \Delta SRF_{p-t} \leq (\Delta SRF_{p-t})_{\min} \end{array}$	Formalin is detected Re-evaluate Re-evaluate Free Probe

Table 4
Arrangement of sensitivity corresponding to sensing layer refractive index from 1.34 to 1.41 for seven different structures at the optimum thickness of TiO2, SiO2 and monolayer of MoS2 and graphene.

Structure configuration	Sensitivity (s) [%RIU ⁻¹]							
	$n_s^2 = 1.34$	$n_s^2 = 1.35$	$n_s^2 = 1.36$	$n_s^2 = 1.37$	$n_s^2 = 1.38$	$n_s^2 = 1.39$	$n_s^2 = 1.40$	$n_s^2 = 1.41$
Conventional	70.00	70.50	71.33	72.00	72.80	73.67	74.42	75.375
Conventional with Graphene	71.00	71.50	72.33	73.00	73.80	74.67	75.43	76.375
Conventional with MoS2	76.00	75.50	76.33	77.25	78.20	79.167	80.286	81.375
Conventional with Graphene-MoS2	76.50	77.00	77.67	78.50	79.40	80.50	81.43	82.625
Conventional with Graphene-MoS2-TiO2	77.00	78.00	79.00	80.25	81.20	82.50	83.71	85.00
Conventional with Graphene-MoS2-SiO2	76.00	75.90	77.53	78.25	79.00	80.00	81.00	82.00
Conventional with Graphene-MoS2-TiO2-SiO2 (Proposed)	78.00	78.50	79.67	80.50	81.80	82.83	84.00	85.375

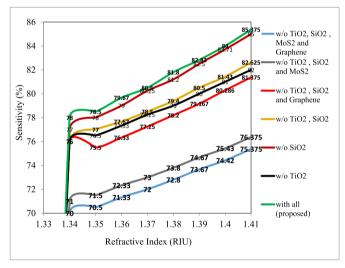


Fig. 4. Sensitivity [%] vs. Refractive Index [RIU] Graph for Different Layer Structure.

The SPR angle increases with the increment of refractive index according to Eq. (1). The variation of sensitivity of the proposed biosensor with respect to the increment of refractive index with a RI step $\Delta n=0.01\ RIU$ is measured and tabulated in Table 4 and it corresponding results is graphically shown in Fig. 4.

We compare our sensitivity analysis among different sensor structure for 1.39 RIU refractive index which is the changed RI of sensing medium after adsorbing 1000 nM perfectly matched target DNA molecule. From Fig. 4, it can easily be observed that the sensitivity without ${\rm TiO_2}$, ${\rm SiO_2}$, ${\rm MoS2}$ and ${\rm Graphene}$ (conventional structure) is very poor and 73.67% whereas the sensitivity with graphene but without ${\rm TiO_2}$, ${\rm SiO_2}$ and ${\rm MoS_2}$ layers is 74.67% consistently better than the conventional structure. This is due to the electron loss of graphene, which is accompanying with the imaginary dielectric constant. This increased SPR angle will lead to obtain increased sensitivity of the sensor as sensitivity is directly related to the variation of SPR angle discussed in Ref. [22]. Furthermore, the sensitivity without ${\rm TiO_2}$, ${\rm SiO_2}$ and graphene but with ${\rm MoS_2}$ layer is 79.167%. Because of ${\rm MoS_2}$'s larger band gap [23], higher optical absorption efficiency [24,25] and larger work

function (5.1 eV) as equated with graphene [26]. The sensitivity of the quantum-confinement-incurred direct band gap in MoS2, allows the high sensitive detection of bio targets. It also holds high fluorescence quenching ability and different affinity in the direction of bio targets [27,28]. After more if both graphene and MoS2 are used and TiO2 and SiO₂ layers are not used then sensitivity improves to 80.5%. This greater than before performance is due to the absorption ability and optical characteristics of graphene biomolecules and high fluorescence quenching ability of MoS₂. Further again, if TiO₂-SiO₂ composite layer is used with the Graphene & MoS2 then the sensitivity enhances from 80.5% to 82.5%. Titanium dioxide (TiO2) and SiO2 have purely real refractive index; hence, can be used as adherence layer above the prism base. As an adherence layer, the composite layer performs better than the individual TiO2 and SiO2 [29,30] because rich plasmon happens at the TiO₂-SiO₂ interface [31]. And this plasmon enhances light trapping effectively [32] which will generate more surface plasmons (SPs). Due to this more surface plasmons (SPs), which will enhance the SPR angle. This increase in SPR angle will increase the SPR sensitivity. Finally the sensitivity for the proposed structure has been carried out, and result is 82.83%, the highest value among all the previous structures. In order to assimilate the advantages of graphene, MoS2, TiO2 and SiO2, we are motivated to use all of them in a single biosensor, which increases the sensitivity.

Lastly, we feel like making a table showing a comparison of sensitivity of the proposed SPR sensor with other existing. Table 5 has been made with taking into account of sensitivity, on the basis of Structure configuration, and operating wavelength sensors in the literature.

4. Conclusion

In this work, a numerical analysis is reported to observe the effect of adding of graphene, MoS_2 , TiO_2 and SiO_2 layer step by step on sensitivity parameters for formalin detection.

The first feature of this study is to detect the presence the formalin based on attenuated total reflection (ATR) method by observing the change of "surface plasmon resonance (SPR) angle versus the change of minimum reflectance" attributor and "the surface plasmon resonance frequency (SPRF) versus maximum transmittance" attributor. Here, Chitosan is used as probe legend to perform particular reaction with the formalin (formaldehyde) as target legend. The second principle feature of this SPR biosensor is the use of graphene, MoS2, TiO2 and SiO2 to

Table 5Sensitivity of the proposed work and comparison with other existing works.

Structure configuration	Sensitivity [Deg-RIU ⁻¹]	Wavelength (nm)	Reference
Conventional with Graphene-MoS2-TiO2-SiO2	82.83	633	In this study
Graphene Coating	57.14	633	[33]
Au-Aluminum thin coating	9.56	680	[34]
Graphene-Ag-Chromium substrate coating	68.03	633	[35]
Au-Ag coating	54.84	632.8	[36]
Graphene Coating	33.98	633	[37]
Graphene Coating with chaclogenide prism	38.88	633	[38]
Transition metal dichalcogenide with silicon nanosheet	89.98	1024	[39]

enhance the sensitivity. The proposed biosensor has a greater sensitivity of 82.83 Deg-RIU⁻¹ as compared to the other reported conventional SPR biosensor. Hence, for the first time as per the best of our knowledge, numerical analysis of Graphene-MoS₂-Au—TiO₂-SiO₂ pentasite layer in a single SPR biosensor is proposed.

Conflicts of interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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