Original Research Article

Sensor Responses of Silver Based Plasmonic Nanoparticles Deposited on PS/P2VP

Abstract

The silver nano particles (Ag NPs) are characterized by modular and unparalleled plasmonic properties. The localized surface plasmon resonance (LSPR) property and their response to the local environment is decisive upon the size and shape of these particles. The LSPR property of nanoparticles has interdisciplinary applications in areas like chemistry, biology and material science with maximum utilization in optical, chemical and biological sensing applications. In this paper, a polymer system is used by blending of two polymers namely, polystyrene/poly (2-vinylpyridine) (PS/P2VP) (50:50) with the optimization technique. Silver nano particles of thickness100, 150 and 200 nm deposited on (PS/P2VP) (50:50) were explored to observe the optical sensitivity. The size and distribution of nanoparticles in the matrices is affected by the nature of polymer composite embedded with silver nano particles. The polymer composites utilized in this study are found to have uniform distribution of nano particles of various sizes.Owing to the unique properties of silver nanoparticles and in order to develop latest plasmonic applications, the optical properties of the nanoparticles embedded in suitable polymer composite were explored. The size of the nanoparticle embedded into a particular polymer composite is the decisive factor for the sensing capability.

Keywords: silver nano particles; polymer composite; thin films; vapor deposition; biosensors

1. INTRODUCTION

Pyridine-containing polymers are water-soluble and coordination reagents for transition metals. Recent time these polymers got attention due to their such applications.The formation of silver particulate films on polymer composite like poly (2-vinylpyridine) yield smaller particles (~ of the order of a few tens of nm) with smaller inter-particle separation. Hence, these silver nano composites are desirable for applications in various fields like drug delivery, antimicrobial activity, wound dressing, catalysis, optical data storage and sensors. The polymer matrix embedded with nano silver particles manifest plasmon resonance absorption resulting in absorption maxima in the visible-infrared region. The parameters like size, shape, filling factor etc of the particle in the polymer matrix decide the spectral position.The surface plasmon resonance absorption for embedded silver in polymer matrix, generally, occurs at a wavelength of ~ 430 nm [1]. This shift of peak towards the higher order of wavelength in the plasmon resonance is due to proximity of the silver clusters [2, 3, 4, 5]. Silver nano particulate films on polymer composite show extraordinary optical properties emerging from the characteristic surface plasmon due to the collective motion of electrons [4,5].The size, shape and dielectric properties of the particle and surrounding medium decide the spectral position, half width and intensity of the plasmon resonance peak [6]. Therefore, nature of metal and surrounding dielectric medium together are important parameters in the excitation of particle plasmon resonance (PPR) and can be manipulated to various applications [7].

Silver, being highly conductive and thermally stable among all metals is considered best for producing nanoparticles and is also considered as an important material in plasmonics [8].

Silver nanoparticles have unique catalytic, electrical, thermal, optical and sensing properties making it suitable for biomolecule detection, immunoassays, surface plasmon optics, data storage, catalysis, surface enhanced Raman scattering, antibacterial material, photonics and photography [9]. The small particles of silver are unstable and tend to agglomerate in porous medium; also, they are toxic to human cells and environment. These effects can be reduced by immersing or encapsulating these nanoparticles in polymer matrices [10, 11, 12, 13]. By doing these the optical, thermal, mechanical and conducting properties of these particles improves making it useful in optical and sensing devices [14].

We analyze polymer composites with applications in sensors. The size of silver clusters in a particular composite is adjusted by varying the thickness of silver film formed on the polymer blend which produces different sizes of nanoparticles. Due to tendency of small silver particles to agglomerate, homogeneous dispersion into the host polymer matrix is not possible by ex situ methods. Moreover, preparation of small silver particles with varying shape and size in different polymer is done by different methods employing potentially hazardous reactants so; to avoid this eco-friendly technique to prepare small silver particles can be used. For reference, vacuum evaporation of silver on softened polymer composites. The softening of polymers is used in order to control, allowing for the embedding of small silver particles into the polymer composites. This forms island and discontinuous silver particulate films by stopping the deposition at early stage. However, due to mobility of islands and coalescence, these films show temporal instability [15] and get oxidized upon when exposed to atmosphere exhibit irreversible increase in electrical resistance [16]. The softened polymer composites are employed for deposition of various inorganic materials, can be referred in [17, 18, 1].

There are limited applications of Ag NPs for biosensors in the literature as compared to gold nano particles (Au NPs). Research on plasmonic biosensors using Ag NPs is even more scarce.While most of the sensors operate ex VIVO, the toxicity of AgNPs is a major concern. Another limitation in using these particles in bare form for biosensing is poor stability and unusual surface chemistry [19, 20].To address such constraints, Ag NPs is coated with large variety of compounds toimprove stability and reduce toxicity of Ag NPs in a given environment [19,21].Additionally, by coating, the aggregation of NPs can be overcomewhich introduces electrostatic, steric, or electrosteric repulsive forces between the particles [19].Various coating methods have been reported for coating Ag NPs with organic or inorganic medium for applications in plasmonic bio-sensing. Embedding silver into polymer matrices is a successfulapproach where silver nano particles are dispersed uniformly in polymer composite preventing agglomeration of nano particles. The optical properties NPs are influencedby the nature of coatingmaterial used and its thickness. The unique characteristic of this hybrid quanta system is largely utilized in plasmonic systems and devices. Optical sensors have applications including refractive index measurement in biomedical, chemical and food processing industries. Consequently, the materials scientists are actively working in the development of ultrasensitive plasmonic detectors for bio sensing applications [22, 23].The present study investigates plasmonic biosensing response of Ag NPs embedded in polymer composites thereby improving electrostatic, steric and electrosteric stabilization of Ag NPs.

This paper is further reporting of work published before for silver nanoparticles deposited on PS/P4VP. The following section 2 gives a peek into the experimental characterization and fabrication of the silver nanoparticles and their deposition into PS/P2VP. In section 3, we briefly discuss the computational software used and step by step method to calculate the various

quantities of interest. Detailed discussion of the results covered in section 4 followed by summary and conclusion in section 5.

2. EXPERIMENTAL STUDY

The structure of Poly (2-vinyl pyridine), P2VP and Polystyrene, PS are shown in figure1 (a) and (b), respectively.

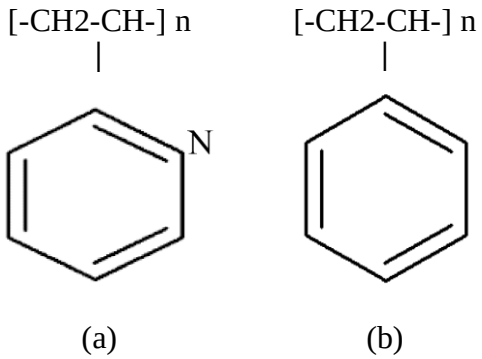

Figure 1: Structure of (a) P2VP and (b) PS

Various polymer compositions are prepared by blending these in solution using dimethylformamide (DMF) as common solvent. Composite of polystyrene/poly (2-vinylpyridine) are prepared in three compositions as {PS (w)/P2VP (w) = 0:100; 50:50; 100:0}. Further, the deposition of silver onto polymer systems is carried out as described in our paper [13].

The particles size of silver clusters embedded inPS/P2VP blends were measured from SEM and XRD from our paper [24] to understand the sensor response discussed in next section.

3. COMPUTATIONAL STUDY

The optical properties of best samples of PS/P2VP (50:50) for 100, 150 &200 nm [24] are used and their sensor responses are recorded using nano sensor lab software. This computational software is based on Mie Theory and is used for investigating the optical response of low-dimensional structures for core-shell nano particle and nano matryoshka; the extinction, scattering and absorption efficiencies. It is designed for a refractive index sensor to simulate sensitivity, figure of merit, quality factor, FWHM (full width at half maximum frequency) and scattering of electromagnetic radiation by core shell spherical nano particles. It is a virtual laboratory with friendly graphical user interface (GUI), an optimization algorithm (to map simulations on experimental results) and scripting capabilities. From our previous morphological studies of silver nano particle composites into above polymer composites, it was found thatthe sizes of nano particles are almost spherical. Therefore, design considerations for spherical nano particle-based simulator panel are used. The scattering, absorption and extension efficiency are calculated by using a flow chart and following parameters.

1) In geometrical panel, the average size of nano particle is taken from the previous study on morphology [24], for example for thickness of 150nm silver on PS/P2VP (50:50); R1=100 nm.

2) Wavelength range: min-400 nm and max-1200 nm and steps 1000.

3) In the material section, import Ag data and plot.

4) Sensing parameter range: n= 1.33-1.37.

5) Select the computerbutton in study section.

6) Graphical results are displayed.

1. RESULTS AND DISCUSSION

There have been studies on silver nanoparticles embedded in polymer composites as humidity sensors [23, 26]. An organic/inorganic composite of poly (diphenylamine sulfonic acid) (PSDA), 3-mercaptopropyltrimethoxysilane (MPTMS), and nano-ZnO has been utilized for constructing thin-film humidity sensors. The humidity sensing characteristics of the sensorare examined through impedance measurements in frequency range 100 Hz - 1 kHz. The sensitivity of the samples increases threefold as significant changes in impedance, along with excellent repeatability and stability across a relative humidity range of 12 - 95% RH is observed [23]. In another study Ag/polymer nano composite is synthesized by a chemical reduction process. The sensing characteristics are examined by forming coatings onto platinum interdigitated electrodes. The sensor provides a reversible, selective, and rapid response proportional to humidity levels in the range of 10-60% RH [26].

Polymer composites that are either grafted or adsorbed with nanoparticles (NPs) promote uniform distribution of NPs when embedded into polymer matrix. The filling of silver onto a polymer composite with essential composition can yield an effective optical sensor. A study on the morphology of Ag NPs embedded in polymer composites, PS/P2VP (50:50), demonstrated homogeneous dispersion of Ag NPs at thicknesses of 100 nm, 150 nm and 200 nm respectively The size distribution and dispersion of the silver nanoparticles depends on the characteristics of the polymer composite and the amount of silver deposited [24]. Embedding Ag-NPs in polymer composites enhances their properties creating a platform for sensor applications due to their large surface area and small dimensions [13, 14]. A variety of nanocomposites with different amounts of Ag NPs can be investigated for the development of efficient sensors and biosensors using nano polymer composites [8].

To fabricate and commercialize highly selective and specific agents, the impact of Ag NPS on parameters such as shape and composition that influence the sensitivity and selectivity of the sensor needs to be analyzed. The extinction efficiencies(Q_{ext}), scattering efficiencies (Q_{sca}), and absorption efficiencies (Q_{abs}) are calculated to examine sensing in Ag NPs. Mathematically they are written as,

$$Q_{ext} = \frac{2}{x^2}\sum_{l=1}^{\infty}[2l+1]Re(a_1 + b_1)$$

$$Q_{sca} = \frac{2}{x^2}\sum_{l=1}^{\infty}[2l+1](|a_1|^2 + b_1|^2)$$

$$Q_{abs} = Q_{ext} - Q_{sca}$$

Where, $a_n's$ and $b_n's$ and defined as;

$$a_n = \frac{\psi_n(x_3)H_n^a(\mu_3 x_3) - \mu_3 Z_n^{(1)}(x_3)}{\xi_n(x_3)H_n^a(\mu_3 x_3) - \mu_3 Z_n^{(3)}(x_3)}$$

$$b_n = \frac{\psi_n(x_3)H_n^b(\mu_3 x_3) - Z_n^{(1)}(x_3)}{\xi_n(x_3)H_n^b(\mu_3 x_3) - \mu_3 Z_n^{(3)}(x_3)}$$

where, μ_1 and μ_2 are the refractive indices of core and the shell respectively, and $\mu_3 = \left(\frac{\varepsilon_3}{\varepsilon_4}\right)$ is the relative refractive index of outer shell medium relative to surrounding medium, $x_1 = kR_1$, $x_2 = kR_2$ and $x_3 = kR_3$are size parameters and, *Riccati*-Bessel functions $\Psi_n(x) = xj_n(x)$, $\chi_n(x) = -xy_n(x)\xi_n(x) = xh_n^1(x)$ of first kind and the logarithmic derivative of *Riccati*-Bessel function like, $Z_n^{(1)}$,$Z_n^{(2)}$, $Z_n^{(3)}$ are written as $Z_n^{(1)} = \Psi'_n(x)/\Psi_n(x)$, $Z_n^{(2)} = \chi'_n(x)/\chi_n(x)$, $Z_n^{(3)} = \xi'_n(x)/\xi_n(x)$ and $H_n^a(\mu_3 x_3)$, $H_n^b(\mu_3 x_3)$.

Themostrelevant parameters in context with sensingare:

1) Qualityfactor(*QF*)ofscatteringresonancepeak.
2) Sensitivity(*S*)
3) Figureof merit(*FOM*).

Thequalityfactorofresonantpeakisdefinedastheratioofresonantwavelengthofpeaktothefull width at half maximum (FWHM) of the resonant peak as [27]

$$QF = \frac{\lambda_R}{FWHM};$$ where, λ_R is the resonant peak wavelength

This suggests that the resonant peak with smaller *FWHM* is associated with higher quality factor. The sensitivity *(S)* of the sensor is defined as the rate of shift of resonant peak wavelength, λ_R, with the variation in the refractive index *(n)* of surrounding medium, i.e.

$$S = \frac{d\lambda_R}{dn}$$

The *FOM* of the sensor is directly proportional to the quality factor of the resonantpeak and the sensitivity,

$$FOM = QF \times S = \frac{\lambda R}{FWHM} \times \frac{d\lambda_R}{dn}$$

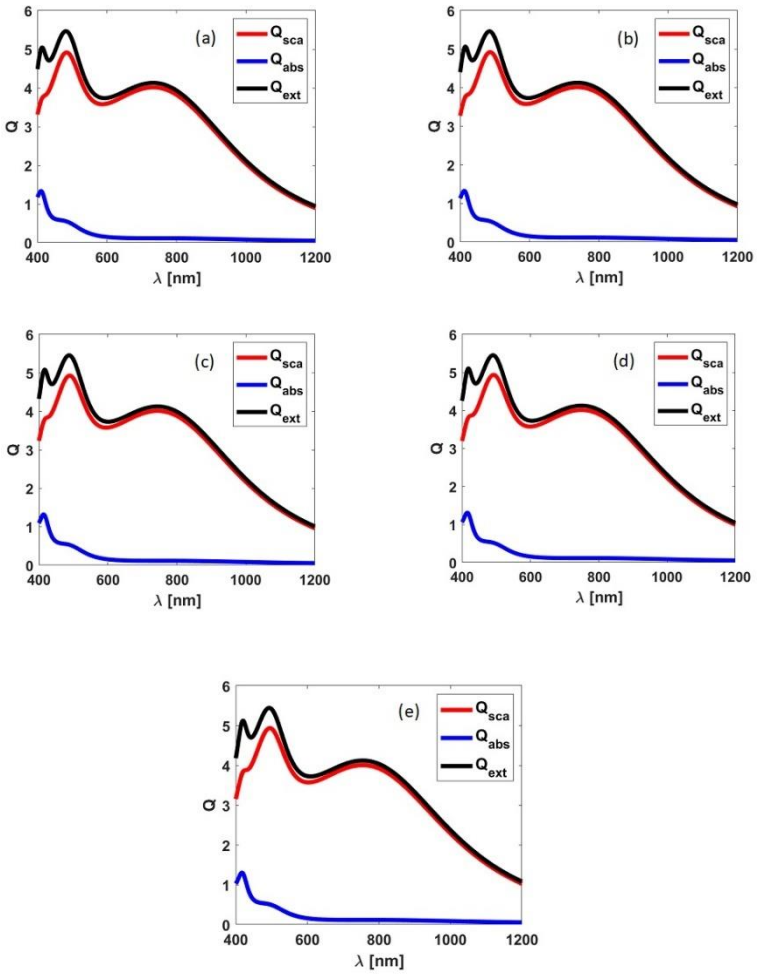

Figure 2: plots of Q_{sca},Q_{abs} andQ_{ext}versus wavelength for different refractive index (a) n=1.33 (b) n=1.34 (c) n=1.35 (d) n=1.36 (e) n=1.37 for PS/P2VP 50:50 at 50 nm

Figures 2-4 show Q_{sca} (scattering efficiency), Q_{abs} (absorption efficiency) and Q_{ext} (extinction efficiency) versus λ (resonance wavelength) for PS/P2VP (50:50) at the thickness 100, 150 and 200 nm, respectively for the various values of refractive indices. It is observed that the nature of graphs is same for refractive indices. Further, two peak resonance wavelengths are found at thickness of 100nm, and onset of a third peak is observed at thickness of 150 nm while distinct three peak resonance wavelengths are observed at thickness of 200 nm. The additional peak wavelength can

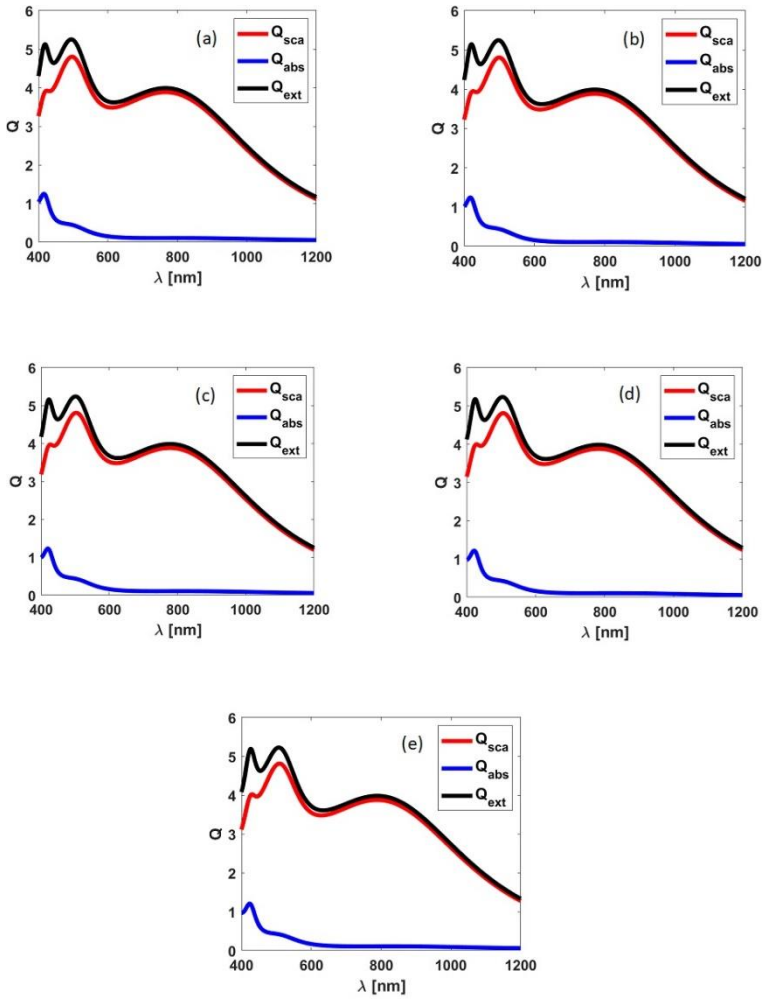

Figure 3: plots of Q_{sca},Q_{abs} andQ_{ext}versus wavelength for different refractive index (a) n=1.33 (b) n=1.34 (c) n=1.35 (d) n=1.36 (e) n=1.37 for PS/P2VP 50:50 at 150 nm

be attributed to the increase in the size of nanoparticles. As the size of the nanoparticles increases, changes in the electron confinement and surface plasmon resonance effects happen which leads to phenomena such as red shift in absorption and scattering spectra. This shift suggests enhanced interactions with light, resulting in alteration in absorption characteristics. Now considering the absorption efficiencies, the overall nature of the graph remains

consistent

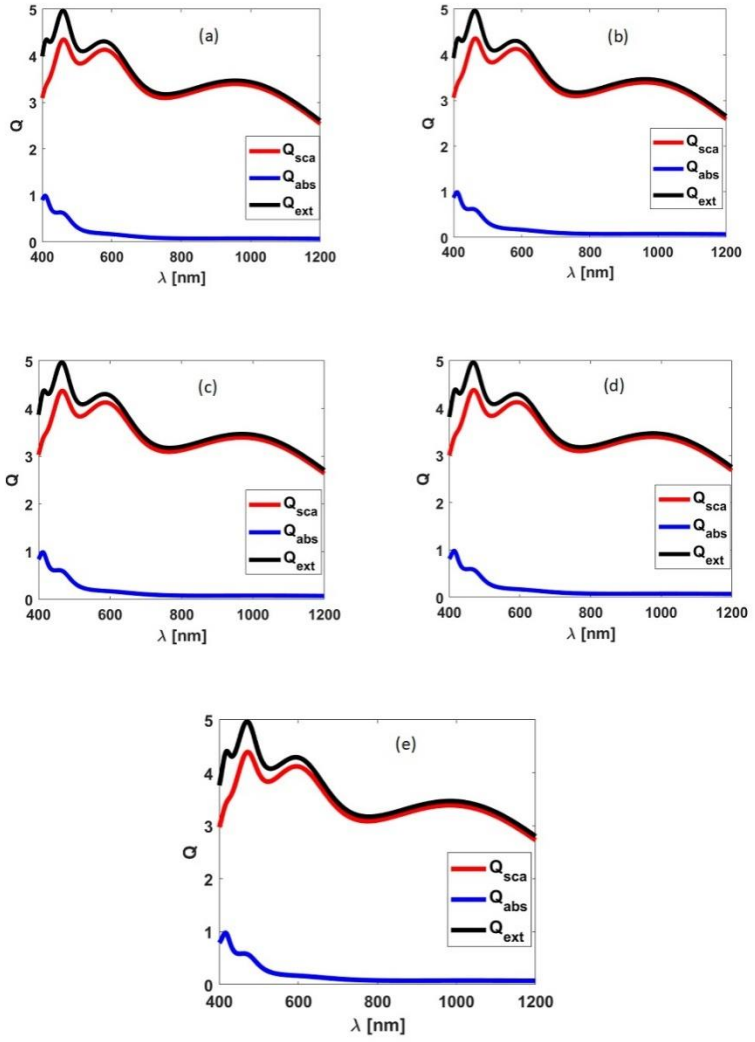

Figure 4: plots of Q_{sca},Q_{abs} andQ_{ext}versus wavelength for different refractive index (a) n=1.33 (b) n=1.34 (c) n=1.35 (d) n=1.36 (e) n=1.37 for PS/P2VP 50:50 at 200 nm

for all refractive indices, however, the peaks are not very clear and sharp for 100 and 150 nm thickness. A peak near 400nm seems to appear and advance towards higher wavelength and disappear with increasing thickness of the NPs. Two distinct peak resonance wavelengths are observed at 200 nm. This can be attributed to the interplay between widened energy levels, shift

in SPR modes, and increased complexity of interactions. Thus, it can be concluded thatall efficiencies show a shift in peak resonance wavelength towards higher wavelength with increase in thickness.

Tables 1-3 presents calculated values of resonant wavelength (λ_R), scattering efficiency Q_{sca}, full width at half maximum (FWHM), quality factor (QF), sensitivity (S) and figure of merit (FOM) for all the samples at different values of refractive index (n). The resonance wavelength, λ_R is measured from the graph obtained by the nanosensor software. This database is useful in choosing a particular optical sensor with requisite peak resonance wavelength. From the observations it is evident that for sensing medium, the size of silver cluster is responsible and not the embedding polymer composite matrix.

Table 1. Sensing performance parameters for PS:P2VP (50:50) (organized with the thickness of 100 nm of silver particulate film). The system is embedded with silver clusters in the polymer composite. Average diameter of nano particle is94.8 nm in the sensing medium range 1.33–1.37.

Sensing medium	**1.33**	**1.34**	**1.35**	**1.36**	**1.37**
λ_r (nm)	733.133	738.739	743.744	749.149	754.755
Q_{sca}	4.914	4.919	4.925	4.93	4.934
FWHM(nm)	76.1	76.9	77.6	76.8	79.2
S (nm/RIU)	560.6	500.5	540.5	560.6	540.55
QF	9.63	9.61	9.58	9.75	9.53
FOM (nm/RIU)	5400.71	4808.05	5180.33	5468.40	5151.30

Table 2. Sensing performance parameters for PS:P2VP (50:50) (organized with the thickness of 150 nm of silver particulate film). The system is embedded with silver clusters in the polymer composite. Average diameter of nano particle is100 nm in the sensing medium range 1.33–1.37.

Parameters/Sensing medium	**1.33**	**1.34**	**1.35**	**1.36**	**1.37**
λ_r (nm)	765.965	771.572	777.177	782.783	788.388
Q_{sca}	4.8	4.803	4.806	4.8086	4.8108
FWHM(nm)	64.8	65.4	66.2	66	64.8
S (nm/RIU)	560.7	560.5	560.6	560.5	560.575
QF	11.82	11.80	11.74	11.86	12.17
FOM (nm/RIU)	765.97	771.57	777.18	782.78	788.39

Table 3. Sensing performance parameters for PS:P2VP (50:50) (organized with the thickness of 200 nm of silver particulate film). The system is embedded with silver clusters in the polymer composite. Average diameter of nano particle is129 nm in the sensing medium range 1.33–1.37.

Parameters/Sensing medium	**1.33**	**1.34**	**1.35**	**1.36**	**1.37**
λ_r (nm)	954.955	962.162	969.369	976.577	983.784
Q_{sca}	4.35	4.362	4.373	4.383	4.393
FWHM(nm)	40.1	39.3	41.7	39.2	40.8
S (nm/RIU)	720.7	720.7	720.8	720.7	720.725
QF	23.81	24.48	23.25	24.91	24.11
FOM (nm/RIU)	17162.99	17644.53	16755.90	17954.57	17378.38

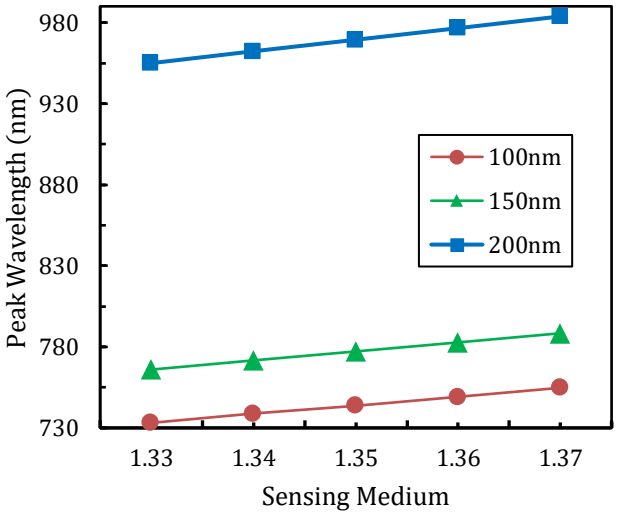

Figure 5.Peak wavelengthvs.Sensing media for PS/P2VP (50:50)

Figure 5 shows the variation of peak resonance wavelength (λ_R) versus sensing medium (n) for all the chosen samples of PS/P2VP with different thicknesses. It is observed that $\lambda_R$shows direct relation with the refractive index of the sensing medium for all samples. It is also observed that the value of λ_R is higher for sample at the thickness of 200 nm which signifies the importance of size of NPs irrespective of polymer composition of matrix.

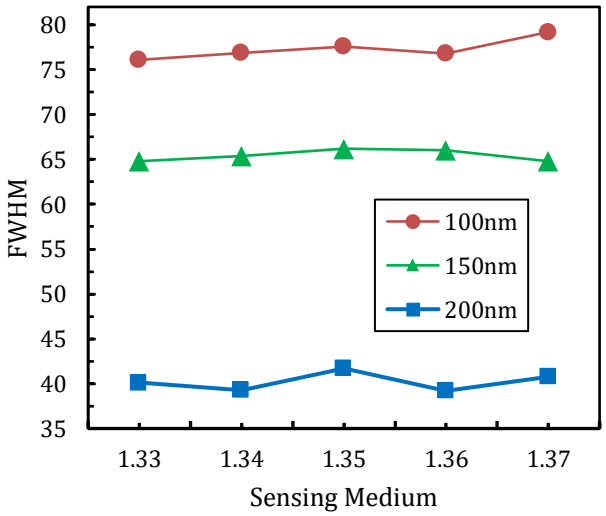

Figure 6. Scattering efficiency vs. Sensing media for PS/P2VP (50:50)

Figure 6 displays the full width at half maximum (FWHM) of the resonant peak plotted against the sensing medium. FWHM does not show much variation by changing the values of sensing medium. However, it is highest for lowest value of thickness.

Figure 7 shows the scattering efficiency of polymer composite embedded with silver nano particles versus sensing medium. The efficiency of all the samples increases with the increase in refractive index of the sensing medium. Here also is quite clear that size of the silver cluster is decisive factor for scattering efficiency of sample. The scattering efficiency is lowest for the nano particles embedded in PS/P2VP for highest thickness.

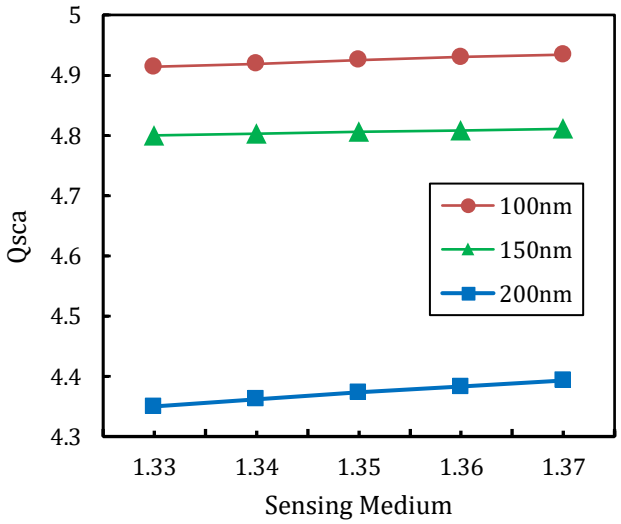

Figure 7. Scattering efficiency vs. Sensing media for PS/P2VP (50:50)

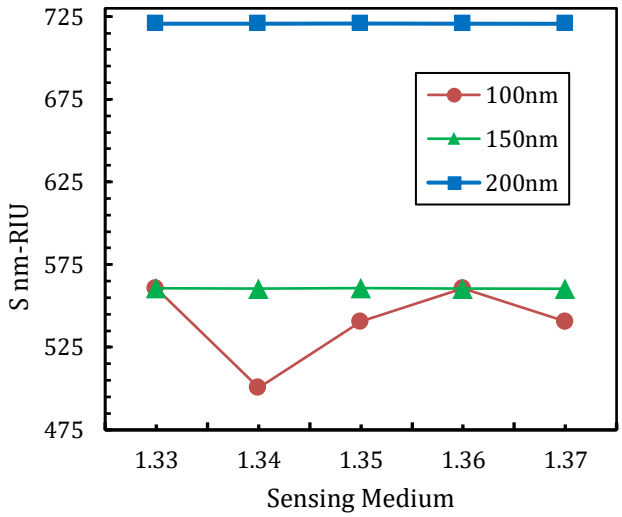

Figure 8. Sensitivity (S) vs. Sensing medium for PS/P2VP (50:50)

Figure 8 shows the variation of sensitivity, S versus sensing medium for all the samples. It is visible that sensitivity is almost constant for PS/P2VP 200nm and 150nm. For lowest thickness there is a variation in the sensitivity with sensing medium.

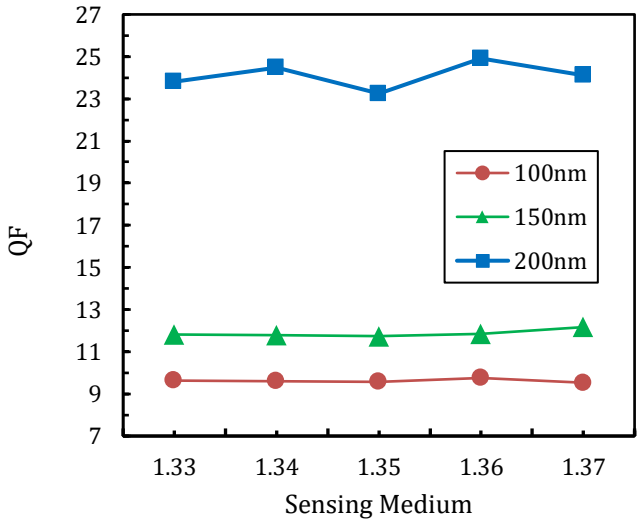

Figure 9. Quality Factor (QF) vs. Sensing medium for PS/P2VP (50:50)

Figure 9 displays the quality factor versus sensing medium for all the samples. QF does not show much variation by changing the values of sensing medium. However, it is highest for highest value of thickness.

CONCULSION

The optical sensor characteristics of silver nano polymer composites based on refractive index measurements have yielded positive results. These silver nano composites were prepared by vapor deposition of silver on softened polymer composites kept in vacuum of the order of 10^{-6} Torr. Previous studies have provided the optimal silver nano composite where silver nano particles are uniformly dispersed with nearly identical size. The sensing properties appear to be dependent on size distribution and dispersion of Ag-NPswithin polymer matrices. Consequently, the samples used here are anticipated to exhibit better sensing properties. The database for the sensing parameters has been compiled in a table for future reference. Additionally, these samples are prepared in thin film form, making them suitable for real time application. In all cases of efficiency, the peak resonance wavelength shifts towards higher wavelength in all the samples for each value of refractive index. Tables 1–3 provide a database tailored for specific application of nano silver polymer composites.

Research Data Policy and Data Availability Statements

All data generated or analyzed during this study are included in this published article [and its supplementary information files].

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