Computational Modelling of Doubly-Photopolymerized Holographic Biosensors

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Holographic sensors are optical devices capable of tuning reflection wavelength, dependent upon nanostructured variations in refractive index. Computational modelling is utilized to simulate the recording and swelling characteristics of developed doubly photopolymerized (DP) holographic sensors. The holographic devices simplify fabrication processes, reduce financial costs, and improve biocompatibility. A holographic grating is achieved through in situ photopolymerization of a highly crosslinked polymer to produce nanostructured refractive index modulation. The unique swelling characteristics DP holographic sensors possess necessitate the development of system-specific computational modelling. Hydrogel parameters, including film thickness, refractive index change, layer number, and external medium refractive index are examined for their effect on reflection spectra. Optimized computational models are utilized to study the effect of differential swelling rates of individual layer spacings on sensor response, indicating an idealized reduction in a swelling of 50% for the highly crosslinked region. A 2D photonic crystal geometry with additional periodicity is developed, to inform further sensor design opportunities. Optimized parameters for both 1D and 2D photonic structures will assist the further development of DP holographic sensors.

1. Introduction

Throughout the COVID-19 pandemic, the importance of rapid medical diagnosis has been paramount, with prompt testing and action for positive cases directly mitigating the health risk to the general population.[1] Point-of-care (POC) diagnostics are key in these situations, through decentralized, fast, sensitive, and cost-effective attributes, which have the potential to revolutionize the approach to medical data collection. The pandemic catalyzed healthcare providers to realize the potential therapeutic improvements offered by POC devices.[2] The POC market is expected to increase 7.3% annually reaching $23.36 billion by 2027.[3] POC devices have previously been developed utilizing electrochemical and fluorescent sensors due to their rapid response time and simple fabrication techniques; however issues such as signal drift, complex readouts, and lack of selectivity have hindered widespread use.[4] Holographic sensors are ideally placed to facilitate the progression towards POC devices, due to their real-time, label-free read-out, low cost, and reversibility.[5] Recent holographic technologies show a primary focus to biomedical applications,[6] such as sensing pH,[7] heavy metals,[8] glucose,[9] ions,[10] and lactate.[11] Holographic recordings can be constructed with additional information such as text to convey further information to users offering both qualitative visual analysis and quantitative measurements through the combination with smartphone applications.[12]

Holographic sensor fabrication techniques and readouts vary significantly depending on the applications and analytes. Common examples are surface relief gratings[13] and phase transmission[14] or reflection holograms.[15] Holographic sensors can be fabricated through the physical casting of surface-based gratings,[16] inkjet printing,[17] and photolithography.[18] Phase

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holograms obtain optical functionality through refractive index (RI) variation, via in situ nanoparticle reduction\[19\] or polymerisation of highly crosslinked nano fringes.\[20\] However, the technology has been hindered due to a range of issues, mainly focused on the use of potentially non-biocompatible silver halide nanocrystals (AgBr NC), complex production techniques, and high cost of materials.\[21,22\] DP hydrogel holograms utilize photolithographic in situ polymerisation to generate nanostructured crosslinking density variations in alternating layers with different RI.\[23,24\] The brightness and color of the hologram can be varied by volumetric change in layers spacing via interaction with analytes.\[25\] With exact determination of the nanostructure layer and swelling degree difficult to be achieved experimentally, computational modelling offers a solution to gain a better understanding of sensor response. Computational modelling has the capability to guide the experimental process through simple and rapid optimization, expediting the prototyping process without the requirement for the expenditure of financial and chemical resources.

Computational modelling is a fundamental tool in scientif...
Figure 1. Schematic depicting the key points of the recording simulation. a) Overview of the geometric structure of the model, inset displays the free triangular meshing utilized; b) electric field intensity plot of counter-propagating 355 nm reference and object beams; c) expanded view of the interference pattern; d) graph to show the effective spatial intensity of the propagating waves across the hydrogel film; e) effective RI recorded within the hydrogel film; f) interference pattern observed as the object beam offset to an angle of 5°.

is dependent upon free radical generation by the photolysis of initiator compounds such as 2-hydroxy-2-methylpropiophenone or 2,2-dimethoxy-2-phenylacetophenone.[35] Figure 1c demonstrates the laser energy profile across the film throughout exposure, effectively highlighting the expected gradient of polymerisation. The intensity of laser light is directly correlated with the extent of polymerisation, and therefore the effective change in RI throughout the film. The electric field intensity of the interference between the two counter-propagating beams is displayed in Figure 1d. A sinusoidal pattern is observable, demonstrating the production of nodes and antinodes with an electric field intensity of 3.5 V m⁻¹. Efficiency of photoinitiator photolysis can be affected by parameters such as concentration, oxygen inhibition, and irradiation time.[36] The electric field intensity gradient between maxima/minima suggests that the degree of polymerisation can be increased with lower photolysis activation energy. In development of experimental prepolymer solutions, results indicate that the lowering of photolysis threshold of the initiator compounds could tune the thickness of initial nanostructured layers of the gratings. Figure 1e reports the effective RI change across the film post-polymerisation. A modulation factor of 0.1 was applied relative to a RI value of 1.4, ensuring results showed relevance to the DP system. Both of RM and IL showed thicknesses ranging from 0.048–0.072 μm throughout the film. Experimentally holograms are recorded in an anhydrous state; therefore upon submerging in an aqueous medium, it is expected that both layers will expand due to the high hydrophilicity of the biocompatible polymers utilized. Herein, by observing the position of individual polymerisation points, although expansion will occur throughout the measuring process, the results demonstrate the origin of the individual polymer layers.

In experimental recording, photosensitive hydrogels are positioned at an angle of 5° for laser exposure. The slight angle allows for the separation of diffracted light from the specular reflection of the probing broadband light. To observe the effect of recording angle variation, the object beam was modelled with an orientation angle of 5° respective to the hydrogel surface, and the corresponding interference pattern was recorded (Figure 1f). The generated electric field demonstrates interference planes at an angle of −5° relative to the hydrogel surface, with a sinusoidal intensity pattern. The simulation indicates produced holograms will show the inverse angle of the recording beam. Although this is not vital to produce simple mirror holograms, complex patterned recording templates should be inverted to ensure that the image is not reversed when illuminated. Simulation of the interference pattern of counter-propagating beams at both 0° and 5° has effectively determined the position and thickness of distinct layers within the hydrogel, improving understanding of the fabrication technique.
Figure 2. Setting up of simulations and overview of geometry. a) Flow chart to illustrate the process of model design, parameter setting, and output optimization to obtain viable results; b) i) 3D projection of the holographic grating geometry and the light path throughout the structure, and ii) schematic depicting the overview of the modelling geometry defining specific regions such as RM and IL; sensor output for c) low reflection and d) high reflection.

2.2. 1D Grating Readout Modelling

Improved understanding of the response in the materials is vital to the development of the DP holographic sensors. Holographic sensors containing alternating nanostructured layers with varied RI generate a selective mirror for wavelengths that conform to the Bragg condition. The smart hydrogel materials’ volumetric response to external stimuli introduces variations in the layer spacing, and therefore tunes the wavelength which can be correlated to a quantitative sensing platform. COMSOL Multiphysics Wave Optics module was chosen to study the sensing performance of holographic sensors. Modelling was achieved through the following fundamental steps (Figure 2a). Prior to geometry design, initial parameters including RI of hydrogel and external medium, layer spacing, and layer number were defined at arbitrary values. Geometry design was guided by theory and holographic structures, as it is expected that the recording pattern is analogous. A 3D projection of the light path through the modelling grating is demonstrated in Figure 2b-i. An external medium size was set to a constant value independent of the grating thickness and spacing, to permit sufficient visualization of the reflected or propagating wave. Electromagnetic Waves Frequency Domain (EWFD) physics settings were applied with a wavelength domain study selected. Electromagnetic Waves Frequency Domain (EWFD) is applicable to solving the calculation of time-harmonic electromagnetic field and wavelength domain studies that permit the examination of sensor response over a range of wavelengths, ideal for studying the relationship of film expansion and reflection wavelength.

Application of physics to the model boundaries required the selection of a periodic input and output, Floquet periodic boundary conditions were applied to the lateral exterior boundaries (Figure 2b-ii). These conditions indicated that the external boundary did not interact with the propagating waves, comparable to
experimental conditions. Two internal arrays were input to represent the alternating layers of high and low RI for RM and IL, respectively (Figure 2b-ii). Due to the lack of curved or triangular components, a physics controlled quadratic meshing was selected; however, both quadratic and free-triangular meshing yielded similar results. To study the interaction between the grating and the propagating wave, the electromagnetic field distribution was examined to observe the efficiency of the reflection (Figure 2c,d). Figure 2c illustrates the expected response of the grating when no major interaction occurs, small harmonic interactions are observable, and the electric field across the model is relatively consistent. When fringe spacings of the grating and the propagating wave satisfy the Bragg condition, a reflection interaction is observed (Figure 2d), where minimal propagation occurs past the grating. Therefore, model geometry was established with physics defined and the determination of the holographic sensor reflection interaction was identified.

2.3. Optical Sensor Readout Optimization

The angle of incidence was varied to observe the angular dependency of the fabricated holographic sensors. Angled recording of holographic gratings separates tuned reflections from the specular reflection reducing interference. Bragg’s equation (Equation (1)) demonstrates sensor response is dependent on the angle of incidence.

\[ \lambda = 2n_g d \sin \theta \]  

where \( n_g \) is the effective RI of the medium, \( \lambda \) is the wavelength of light, \( d \) is the grating constant, and \( \theta \) is the diffraction angle. The focus of these simulations is the identification of a potential disparity between simulated planar gratings and the angled gratings expected to be produced experimentally. Angles were varied over a range of 0–25° (Figure 3a). Minimal variation in diffraction efficiency or wavelength occurs for illumination angles of 0–5° (Figure 3b). Upon increasing the incident angle from 10–25°, replay wavelength decreased while diffraction efficiency increased. This phenomenon is conducive as at shorter wavelengths the differences in phase were more significant as RI modulation remained constant. The minimal differentiation of the peak replay wavelength for 0–5° validates that modelled results are viable. In experimental processes, if a reflection angle of more than 5° is necessitated, replay wavelength and efficiency of reflection can vary.

A computational study of key parameters in DP holograms was conducted to simulate their influences on the spectral readout. A range of parameters can influence diffraction efficiency, for example, RI modulation and layer number are identified as key in the simulation process. RI modulation is directly linked to the diffraction efficiency of incident light. The RI polymeric materials generally range from 1.30–1.70. As the hydrogels in RM are comprised predominantly of water due to low crosslinking density and high hydrophilicity, RI for the RM was set to 1.335. The RI of IL is expected to be higher due to the increased crosslinking density (≈1.4–1.65). RI variation upon swelling is expected in response to stimuli. The exact RI of IL remains an unknown parameter as nano fringes are not discernible through experimental optical analysis. Furthermore, the exact degree of mixing/polymerisation is unable to be identified; therefore, the degree of polymerisation and mixing of the two polymeric materials cannot be confidently calculated. To determine the effect of RI variation in IL, a range from 1.35–1.65 was examined, whilst RM RI was fixed at 1.335 (Figure 3c). As RI of the IL layer increases, the diffraction efficiency of reflection peaks and the harmonic peaks increased. This phenomenon is due to the high RI modulation between two layers generating a more efficient holographic structure. When observing the diffraction efficiency change of the main reflection peak (≈400 nm) and the harmonic peak (≈550 nm), two different rates of increase occur (Figure 3d). This can be attributed to the harmonic interactions being more dependent on the RI of IL. As harmonic peaks are rarely observed in holographic devices, it is assumed that the effective RI of the IL layer must be below where the harmonic peak is present. The RI of IL of 1.47 gives the result with the highest diffraction efficiency feasible whilst diminishing the harmonic peak and was therefore taken forward. As it is expected due to the high level of crosslinking density present in the IL layer that swelling will be minimal, the value determined was considered constant throughout the following simulations. A summary of the parameters and characteristics of both RM and IL layers is listed in Table S1, Supporting Information.

Although RI has a significant effect on the ability of holographic sensors to interact with propagating light waves, the thickness of holographic sensors also determines diffraction efficiency. Experimentally, photosensitized hydrogels are dried to a minimal thickness pre-exposure with nodes and antinodes between counter-propagating laser light producing points of polymerisation. Controlling overall hydrogel film thickness regulates the number of recorded layers in the holographic sensor. Thin hydrogel films are ideal for sensors as thicker films hinder the rate of diffusion through polymeric networks reducing response time. The number of layers relates to the individual repeating unit, one pair of RM and IL constitutes a layer. A range of 5–30 layers increasing in an increment of 5 was set (Figure 3e). Diffraction efficiency increases linearly until 15 layers with negligible additional interaction in the remaining section which can be described by the “weak grating approximation” (Equation (2)).

\[ R = \tan h^2 \left( \frac{\pi \Delta n L}{\lambda} \right) \]  

where \( R \) is diffraction efficiency, \( \Delta n \) is the RI modulation, \( L \) is the length of the multilayer Bragg structure, and \( \lambda \) is the wavelength, from this \( \Delta n \) value of 0.003 can be calculated. The result demonstrates a point at which film thickness can be optimized for the highest diffraction efficiency whilst minimizing the negative characteristics of thicker films (Figure 3f). However, it must also be considered that the values are for an optimized RI modulation, with lower RI modulation it is expected that a higher layer number would be required to obtain comparable results. Throughout the following 1D model experiments, 15 layers were carried forward to reduce computation complexity without hindering output. Here, parameters with high diffraction efficiency have been examined and optimized to inform and guide the sensor design process.
Figure 3. Optimization of model analysis parameters; a) surface plots showing the wave propagation with varying angle of incidence for i) 0°, ii) 5°, iii) 10°, iv) 15°, v) 20°, and vi) 25° at minimum (upper) and maximum (lower) interaction, respectively; b) simulated reflection spectra demonstrating the effect of incident angle variation; c) spectral response of IL RI variation over the rational experimental range; d) comparison of the diffraction efficiency between main reflection peaks and the harmonics; e) simulated spectral graph depicting the diffraction efficiency variation of reflection spectra for different layer numbers; f) surface plot with height to demonstrate the wave propagation through the grating for layer numbers 5–30, respectively, inset compares the diffraction efficiency for increasing layer numbers.

2.4.1 DHologram Swelling Optimization

The reflected wavelength is determined by grating periodicity, which can be tuned through the expansion or shrinking of the hydrogel film. Swelling behaviors of the DP sensors have not yet been established in experimental measurements, as the photonic structure is difficult to examine through conventional analytical techniques such as optical microscopes and electron microscopy. Maintaining RI, layer number and incident angle parameters established in previous optimizations, layer spacing was varied to examine the response. Regardless of the variation in crosslinking density across the hydrogel film, both RM and IL are varied at the same rate. Therefore, an initial spacing range of 200–300 nm with an increment of 20 nm was set (Figure 4a). Figure 4b demonstrates the trend in peak variation compared to the expected values dictated by Bragg’s law, alongside the corresponding colorimetric response. Disparity in the simulated wavelengths when compared to the expected values is observed; however, the rate
of wavelength change is comparable to layer spacing. It is plausible experimentally that the value for IL may be below that of RM due to a lack of polymerisation efficiency, for example if the activation energy of photoinitiators is greater than 1/2 the total exposure energy. Therefore, IL was examined over 80 to 105 nm whilst maintaining an RM spacing of 200 nm to obtain an ideal Bragg replay wavelength (Figure 4c). IL shifting displays a linear trend, with an optimum reflection wavelength for IL at 90 nm delivering a response closest to that of the expected wavelength at 400 nm (Figure 4d). The expansion of IL with a high crosslinking density is minimal when introduced to an aqueous environment. Therefore, IL was maintained at the idealized value (90 nm) and RM was varied through a 200–300 nm range (Figure 4e). It was observed that the rate of change in Bragg replay wavelength reduced agreement with theory, indicating that both RM and IL must swell simultaneously to maintain the idealized response (Figure 4f). As the IL is polymerized within the RM, it is expected that an expansive force would be induced due to stimuli-responsive co-monomers. Initial RM and IL values of 200 and 90 nm, respectively, show the strongest correlation with theory; however, a requirement for both layers to swell simultaneously at different rates has been identified.

To examine the optimal layer spacing variations, RM and IL were increased at different rates to reflect expected results more closely. Relative IL expansion values of 25%, 50%, and 75% were set to optimize film response (Figure 5a–c). The RM layer was set to 200, 220, 240, 260, 280, and 300 nm, IL was set to an initial value of 90 nm. The ratioed swelling of the film can facilitate more accurate estimations of the degree to which the IL expands.

Figure 5a highlights the simulated Bragg reflection spectra at a relative expansion of 25%. With RM varied from 200 to 300 nm, IL spacing was set to 90, 95, 100, 105, 110, and 115 nm respectively. Over the IL spacing range of 90–100 nm, diffraction efficiency increased \( \approx 10\% \) and a maximum is attained at an IL spacing higher than 100 nm. Harmonic peaks occur between 350–400 nm, hypothesized to be linked to additional resonances within the structure. The correlation between peak wavelength and layer spacing remains below the predicted values. 50% swelling shows a significantly improved sensing performance (Figure 5b). Throughout the range of expansions, a diffraction efficiency of \( \approx 100\% \) is maintained without harmonics. Figure 5c demonstrates the simulated spectral response at a relative expansion of 75%. Diffraction efficiency fades, comparing the 84% diffraction efficiency at RM = 300 nm of 75% swelling to 25% and 50% where diffraction efficiency is near 100%. IL expansions of 135, 150, and 165 nm harmonic peaks reoccur over the 350–450 nm range invalidating the expansion ratio. Figure 5d illustrates the reflection peak shift for each swelling ratio when com-
Figure 5. Optimization of peak replay wavelength through correlated simultaneous swelling; a–c) simulated spectral response of gratings where IL thickness is varied at a) 25% ($y = 1.6943x + 59.42$), b) 50% ($y = 2.0457x - 10.762$), and c) 75% ($y = 2.3857x - 78.762$) of the rate of IL; d) simulated peak wavelength change compared to the expected wavelength response; e) surface plot of the spectral responses of the three expansions at the expected wavelength of 600 nm; f) simulated reflection spectra of the holographic sensor in a variety of external medium RI.

pared to the expected Bragg’s law response, with 50% showing the highest agreement. Figure 5e demonstrates the electric field surface plot of the maximum expansions of RM at 25%, 50%, and 75% with an expected replay wavelength of 600 nm. It can be observed that the maximum expansion at 50% shows strong interactions with high diffraction efficiency whereas 25% and 75% display weak interactions. The limitation of IL expansion at 50% for RM is consistent with the predicted wavelength spacing relationships. Therefore, it will be carried forward throughout the following modelling studies.

DP holographic sensors have shown the sensitivity to alcohols via the use of poly(hydroxyethyl)methacrylate hydrogel systems. Sensing properties of hydrogels were not affected by minor external RI variation in the presence of alcohols. Future sensors may be capable of sensing analytes with significantly higher RIs. The effect of external RI was studied with values varied from 1.1 to 1.5 to cover a range of feasible potential analytes. Figure 5f highlights that as external RI is increased, the diffraction efficiency of the sensor decreased significantly. As external medium RI increased, the RI modulation between the internal layers is diminished, and therefore decreased the diffraction efficiency of the gratings. Since the developed holographic sensors have been optimized for an aqueous environment, external RI was decreased inducing a rise in diffraction efficiency due to the increase in effective RI modulation relative to the external medium. Simulations should consider the RI of analytes in the sensor design for the application along with functionality. If there is a requirement to utilize sensors in excessively high or low RI environments, the materials utilized should be adjusted accordingly.

2.5. 2D Optical Structure Design and Readout

To enhance the sensing performance of holographic hydrogels through the reduction of highly crosslinked regions, a computational 2D photonic structure was proposed. The increased crosslinking density within DP holograms can affect hologram sensitivity due to a decreased ability to swell, a hypothesis to utilize a 2D photonic structure with periodicity in both of $x$ and $y$ directions was developed. A schematic of the proposed 2D photonic structure is highlighted in Figure 6a. The removal of optical grating sections is expected to impede the response of the holographic sensors alongside the introduction of new optically active structures. Due to periodicity being present on the $x$-axis (RM$_2$/IL$_2$, Figure 6b), a transmission grating interaction can be a potential source of interference. The 2D holographic grating was set to a thickness of 10 layers with a vertical layer number of 7.

Fabrication of RM$_2$/IL$_2$ periodicity can be considered analogous to experimental recording processes using photomasks. To
facilitate experimental measurement, hydrogel sensing films are chemically attached to a glass substrate, this could however produce a high level of restriction in the direction normal to the surface due to few degrees of freedom. Therefore, the expansions of both RM₂ and IL₂ are maintained comparably. To investigate the effect of layer spacing on the diffraction efficiency of the gratings, RM₂ was set to values of 50–100 nm (Figure 6c), whilst maintaining the optimized parameters for spacing and RI for RM₁ (200 nm, 1.335) and IL₁ (90 nm, 1.47). As RM₂ increased from 50–100 nm, a general decrease in the diffraction efficiency from 32% to 26% occurs. Diminishing diffraction efficiency can be linked to γ-axis periodicity that impedes the interaction. As the designed model aims to examine reflections normal to the input port, additional transmission diffraction orders that occur in the γ-axis may not be visible in this model. The decrease in diffraction efficiency does not affect the wavelength of the reflection, indicating that the wavelength shift of the grating is independent. Individually examining the effect of IL₂ highlighted its opposing effect on diffraction efficiency over a parameter range of 50–100 nm, where diffraction efficiency increased from 8% to 18% (Figure 6d). This is intuitive as the proportion of the hydrogel structure containing low RI layers remained constant, whilst the high RI regions are expanding. Minimal diffraction efficiencies of the 2D photonic structure can be overcome experimentally.

Figure 6. 2D photonic crystal modelling. a) 3D projection of proposed 2D structure demonstrating modelled light path. b) Schematic of 2D photonic structure model displaying the additional RM₁-IL₂ spacing parameters. The magnified diagram shows the modelling of structure and meshing. c) Expansion of RM₂ whilst maintaining all other variables. d) Variations of IL₂ whilst maintaining all other variables. e) Variations of RM₁ and IL₁ at a ratio of 2:1 for volume change. f) Variation of RM₁, IL₁, and IL₂ whilst maintaining RM₂. g) Variation of all parameters in a ratioed fashion. h) Electric field spectra of finalized swelling behaviors.
by utilizing a brighter or more intense broadband light source to improve the intensity of observable reflections.

For further understanding of how the additional freedom of movement modifies readout, layer spacing was varied at the previously optimized rate of 2:1 RM/IL, with initial values at 200 and 90 nm respectively, whilst RM parameter values were set to 50–100 nm (Figure 6e). Diminishing of diffraction efficiency associated with the individual expansion of RM was present within the spectra displayed, highlighting that the trend is consistent with previously demonstrated wavelength variation. Figure 6f displays the effect of IL variation set to values of 50–100 nm whilst RM:IL are increased. A continuation of the trend in increasing diffraction efficiency could be observed whilst maintaining consistency in wavelength shift, further confirming the conclusion that diffraction efficiency is determined by the RI modulation throughout the grating whereas wavelength is established through the -axis expansion. Finally, all periodicities are varied simultaneously to simulate a real-world expansion of the 2D holographic grating (Figure 6g). Diffraction efficiency is maintained consistently across the spectra with minimal variation. The equal variation of both RM and IL obtained a readout of continuous diffraction efficiency of reflection, indicating a competing effect. It is hypothesized that the ratio of RM:IL determines the diffraction efficiency. Figure 6h shows the electric field density plot of the respective expansions at the wavelength of highest interaction, where a clear reflection interaction occurs. Simulations of the 2D holographic grating have informed the experimental process of several parameters which need to be maintained to achieve a viable sensor. Replay wavelength is unaffected by additional periodicity in the -axis; however, the diffraction efficiency is reduced. The interference layer reduction may allow for easier expansion of the holographic sensor and facilitate higher sensitivity to analytes.

3. Conclusion

DP holography shows promises to assist in the development of POC devices, due to their rapid, label-free, and low-cost fabrication.[38] Sensors have been simulated to expedite the development of biocompatible POC sensing devices. Computational models were designed utilizing the COMSOL Multiphysics Wave Optics module to study the fundamentals of recording, readout, and optimization within DP holographic sensors. Recording simulations indicate the points of polymerisation throughout the hydrogel structure, facilitating informed assumptions about the internal polymeric network structure. Film thickness and RI variation between interior layers and external medium were found to be significant parameters in correlation with the diffraction efficiency.[39] Sensors with 15 layers with an IL RI of 1.47 demonstrated results that correlated with responses predicted by Bragg law. Studies of the ratioed swelling behaviors within 1D photonic structures demonstrated readouts correlating with theory, assisting in the expansion of the understanding of the response mechanics. Optimized swelling characteristics were identified as an initial spacing of 200 and 90 nm for RM and IL, respectively, following a relative expansion of 2:1 (RM:IL). To advance sensitivity of the holographic sensors, a 2D grating was proposed and optimized to minimize swelling restriction. Future developments of this computational modelling could focus on the use of a sinusoidal RI change within the grating to reflect the experimental structure. The generic model designed herein can be utilized to inform the development of DP devices once stimuli responsive hydrogels have been selected, the incorporation of chemical characteristics such as swelling rate, rate of diffusion, or activation energy of photoinitiator compounds could improve the model’s application to sensor design. Overall, the comprehension of the DP systems characteristics and internal mechanics has been expanded, with the experimental process informed of the key parameters in a sensor design characteristic optimization. Also experimental projects of the advanced modelling developed here can be utilized to highlight the potential routes to improving sensor characteristics.

4. Experimental Section

Computational Simulations: Model design and study execution were conducted on COMSOL Multiphysics 5.6 Wave Optics module based on a Lenovo ThinkPad E14, Intel Core i7 processor, 64-bit, supporting Windows 10. Modelling of holographic recording mechanism was adapted from COMSOL Multiphysics “Single Bit Hologram” tutorial, with parameters and properties optimized to allow for application to a doubly photopolymerized holographic biosensor recording set up. The geometry consisted of a union between a square to represent the external medium (height = 30 μm, thickness = 30 μm), and a rectangle (height = 20 μm, thickness = 10 μm). RI of the external medium and the internal hydrogel structure were set to 1.00 and 1.47, respectively. Ports were positioned at opposing ends of the model, to adequately represent the holographic recording set up utilized for reflection holography. A wavelength of 355 nm was selected to coincide with the recording wavelength utilized in UV photopolymerized holographic sensor fabrication. Induced modulated RI post recording was collected via a 2D cut line through the hydrogel with an inferred exposure threshold of 0.4. Holographic sensor readout models were designed with a fixed size exterior medium positioned at either end of a grating structure. The readout model was designed with positioning of the two exterior mediums (height = 4000 μm, thickness = 4000 μm, RI = 1.33) at points relative to the grating thickness to minimize the risk of overlap. Periodic input and output ports were positioned on opposing vertical faces of the model, with a Floquet periodic condition applied to the lateral boundaries. The grating array was designed with individual parameters and position relative to the opposing layer thickness (height = 3000 μm, thickness = 50–300 μm). Parameters such as RM/IL spacing, refractive indices, and angle of incidence were all set prior to sensor design to arbitrary values to permit optimization of the model. Blank materials were utilized for the interior medium with an RI set relative to the input parameters highlighted throughout the manuscript. An extra fine physics controlled quadratic mesh was selected to ensure the correct resolution and reliability of studies. An electromagnetic wave frequency domain physics package was utilized for the computation of optical response. Parametric sweeps facilitated the study of individual parameters with a wavelength range set to 350–700 nm to correspond with the visible range. Angle of incidence was altered at the incident port, with angles varied via parametric sweep (0–25°). The results of the electric field distribution and reflectance were exported from COMSOL or to Excel to allow for simple processing and analysis.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

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