Encapsulation of Methyl Orange pH-Indicator into a HYPR-Modified Sol-Gel Silica Matrix

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Abstract

The immobilization of pH-sensitive organic dyes within sol–gel silica matrices has attracted considerable attention due to its potential in the development of solid-state optical sensing systems. In the present study, methyl orange (MO) was encapsulated into a HYPR-modified silica matrix prepared through an acid-catalyzed sol–gel process using tetraethyl orthosilicate (TEOS) as the silica precursor. The incorporation of a cationic surfactant during matrix formation was intended to improve the structural characteristics and accessibility of the porous silica network.

The resulting monolithic silica materials were transparent and mechanically stable, demonstrating successful immobilization of methyl orange within the inorganic–organic hybrid framework. Spectroscopic evaluation revealed that the encapsulated indicator preserved its pH-responsive behavior after immobilization, exhibiting optical characteristics comparable to those of the free indicator in solution. Surface morphology analysis suggested a homogeneous distribution of methyl orange molecules within the modified silica structure and indicated weak interactions between the indicator molecules and the host network.

The findings confirm that HYPR-modified sol–gel silica matrices provide a suitable environment for the immobilization of methyl orange while maintaining its sensing functionality. The developed hybrid material shows promising potential for application in solid-state pH monitoring and optical sensing technologies.

Keywords: Methyl orange; Sol–gel silica; HYPR surfactant; Immobilization; Hybrid materials; pH sensing.

1. Introduction

Sol–gel technology has emerged as an attractive approach for the preparation of organic–inorganic hybrid materials owing to its simplicity, versatility, and ability to produce highly porous silica networks with tailored physicochemical properties. These materials have found extensive applications in catalysis, sensing technologies, environmental remediation, and advanced functional materials due to their high surface area, optical transparency, and chemical stability (Hench & West, 1990; Brinker & Scherer, 1989).

Among the various applications of sol–gel materials, the immobilization of pH-sensitive dyes has received considerable attention for the development of solid-state optical sensors. Encapsulation of indicator molecules within silica matrices provides protection against leaching and chemical degradation while maintaining their responsiveness to environmental changes. The porous nature of sol–gel silica networks facilitates mass transfer and allows analytes to interact efficiently with immobilized sensing species (Wolfbeis & Rodriguez, 1992; Lobnik et al., 1998).

Methyl orange (MO) is a widely used azo dye for pH determination because of its distinct color transition and reliable optical response under acidic and alkaline conditions. Previous studies have demonstrated the successful immobilization of organic indicators within sol–gel-derived silica matrices, highlighting the potential of these systems for sensing applications (El-Nahhal et al., 2001; Zaggout et al., 2005).

Recent advances in hybrid sol–gel materials have expanded the range of encapsulated functional molecules and improved the performance of optical sensing platforms. Organic–inorganic hybrid membranes and mesoporous silica systems have shown promising characteristics for pH sensing and dye immobilization applications due to their enhanced structural stability and accessibility of active sites (Ferreira et al., 2022; Alenazi et al., 2024).

Furthermore, surfactant-assisted sol–gel synthesis has been recognized as an effective strategy for controlling pore structure, surface morphology, and molecular distribution within silica networks. In particular, cationic surfactants have been reported to improve the organization of porous frameworks and influence the physicochemical properties of sol–gel-derived materials (Koç Keşir et al., 2022, Tong & Febles 2024). Such modifications can improve the accessibility of immobilized molecules and enhance their interaction with the surrounding environment.

In the present study, methyl orange was encapsulated within a HYPR-modified silica matrix prepared through an acid-catalyzed sol–gel process using tetraethyl orthosilicate (TEOS) as the silica precursor. The optical properties, surface morphology, and pH-responsive behavior of the resulting hybrid material were investigated to evaluate its suitability for solid-state pH sensing applications.

2. Experimental

2.1. Reagents and Materials

Tetraethyl orthosilicate (TEOS) was used as the silica precursor for the preparation of the sol–gel matrix. Methyl orange (MO) was employed as the pH-sensitive indicator, while a HYPR surfactant (Alkyl Dimethyl Hydroxyethyl Ammonium Chloride) was utilized as a structure-modifying agent during the sol–gel synthesis process. Absolute methanol was used as a solvent, and hydrochloric acid (HCl) was employed as the catalyst for hydrolysis and condensation reactions. All chemicals were of analytical grade and were used as received without further purification.

2.2. UV–Visible Spectroscopy

The optical properties of the immobilized methyl orange were investigated using UV–Visible spectroscopy. Spectral measurements were carried out in the wavelength range of 200–800 nm under both acidic and alkaline conditions to evaluate the retention of the indicator's optical response following immobilization within the silica matrix. The spectra were recorded using a SHIMADZU UV–VIS–NIR spectrophotometer (Model UV-3101-PC).

2.3. Surface Morphology Analysis

The surface morphology and topographical characteristics of the prepared silica monoliths were examined using White Light Interferometry (WLI) (Zygo NewView 5000). This non-contact optical technique provides three-dimensional imaging of surface features and enables accurate measurement of surface roughness and microstructural characteristics.

The instrument operates by dividing a white light beam into two optical paths. One portion is reflected from the sample surface, while the other is reflected from a high-quality reference surface. The resulting interference patterns are captured by a solid-state camera and processed using MetroPro software to generate three-dimensional surface maps. The technique allows detailed evaluation of surface texture, pore distribution, waviness, step height, and roughness parameters with high vertical resolution.

The average surface roughness (Ra) was calculated according to Equation (1):

$$Ra = (\Sigma|y_i|)/N$$

where yi represents the absolute deviation of each measured point from the mean surface level and N is the total number of measured points included in the analysis.

2.4. Preparation of Silica Monoliths

The silica monoliths were prepared through an acid-catalyzed sol–gel process. TEOS was mixed with methanol and hydrochloric acid under continuous stirring to obtain a homogeneous sol solution. After complete hydrolysis of the silica precursor, methyl orange was introduced into the reaction mixture followed by the addition of the HYPR surfactant. The resulting solution was stirred thoroughly to ensure uniform distribution of all components within the sol.

The prepared sol was transferred into glass molds and maintained at room temperature until gelation occurred. Following gel formation, the samples were aged under ambient conditions to promote further condensation and structural stabilization of the silica network. Transparent and mechanically stable monolithic silica disks were obtained after the aging process.

2.5. Washing and Drying Procedure

The prepared monolithic disks were washed several times using a methanol/deionized water mixture (1:1, v/v) to remove non-entrapped methyl orange molecules from the silica network. The washing solutions were collected for subsequent analysis of dye leaching. After washing, the monoliths were dried at room temperature until constant weight was achieved and then stored in sealed containers prior to characterization.

2.6. Entrapment Capacity of Methyl Orange

The entrapment efficiency of methyl orange within the silica monoliths was determined by comparing the amount of dye initially introduced into the sol–gel system with the amount released during the washing process. The quantity of entrapped dye was calculated as the difference between the initial amount added and the amount detected in the collected washing solutions.

The entrapment capacity of methyl orange at different initial concentrations is presented in Table 1.

Table 1 Entrapment capacity of methyl orange in silica monoliths at different initial dye concentrations

Original concentration (M)	Calculated mg added	mg leached	mg entrapped	mg entrapped/mg added
0.05×10^{-4}	0.00163	0.00112	0.00051	0.31
0.5×10^{-4}	0.0163	0.0100	0.0062	0.38
1.5×10^{-4}	0.0490	0.0330	0.0160	0.32
5.0×10^{-4}	0.1630	0.1110	0.0520	0.37
50.0×10^{-4}	1.6300	1.1040	0.5260	0.32

3. Results and Discussion

3.1. Synthesis of HYPR-Modified Silica Monoliths

Methyl orange was successfully immobilized within silica monoliths through an acid-catalyzed sol–gel process. Hydrolysis and condensation of tetraethyl orthosilicate (TEOS) produced a transparent silica network capable of accommodating indicator molecules while preserving their optical functionality. The incorporation of the HYPR surfactant contributed to the modification of the silica framework and promoted the formation of a more homogeneous porous structure.

Transparent and crack-free monoliths were obtained after gelation and aging under ambient conditions. The resulting materials exhibited good optical transparency and mechanical stability, indicating successful formation of the hybrid silica matrix and effective incorporation of methyl orange within the host network.

3.2. Entrapment Capacity of Methyl Orange

The entrapment efficiency of methyl orange was evaluated by comparing the amount of dye initially introduced into the sol–gel system with the amount released during the washing process. The experimental results are summarized in Table 1.

The amount of methyl orange retained within the silica matrix increased proportionally with the initial amount added to the precursor solution. As illustrated in Figure 1, a nearly linear relationship was observed between the initial dye content and the amount entrapped within the monolithic silica matrix. This behavior indicates a relatively homogeneous distribution of methyl orange molecules throughout the porous silica network and confirms the effectiveness of the immobilization process.

The results further suggest that the modified silica matrix provides sufficient accessible sites for dye incorporation while minimizing dye loss during washing. The observed trend demonstrates the suitability of the HYPR-modified silica network for immobilization of pH-sensitive organic indicators.

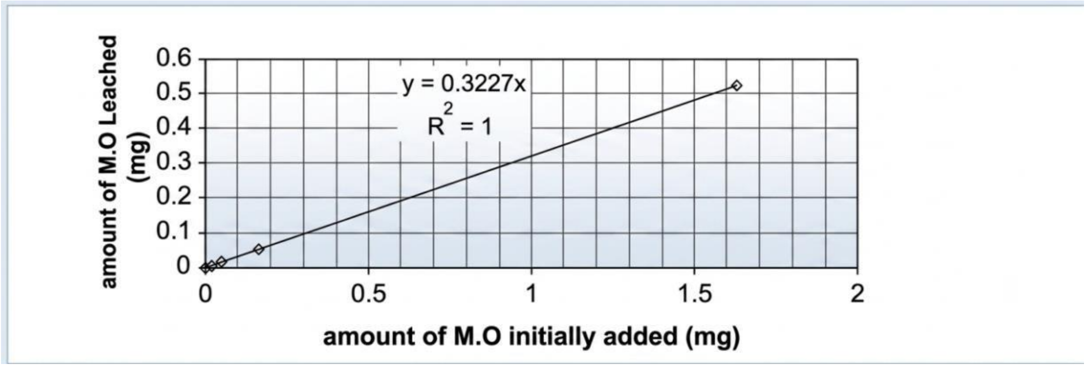

Figure 1. Entrapment of methyl orange in HYPR-modified silica monoliths as a function of the initially added amount.

3.3. UV–Visible Spectral Analysis

The UV–Visible spectra of methyl orange before and after immobilization were recorded to evaluate the preservation of its optical properties following encapsulation within the silica matrix. The comparison between free methyl orange and immobilized methyl orange is presented in Figure 2.

The spectra reveal that the characteristic absorption band of methyl orange was retained after immobilization, indicating that the encapsulation process did not significantly alter the molecular structure of the dye. The similarity between the spectra of free and immobilized methyl orange confirms that the indicator remained chemically stable within the silica framework.

Furthermore, the porous structure generated within the HYPR-modified silica matrix may facilitate interaction between the encapsulated dye molecules and the surrounding medium, thereby improving accessibility and maintaining the sensing functionality of the indicator.

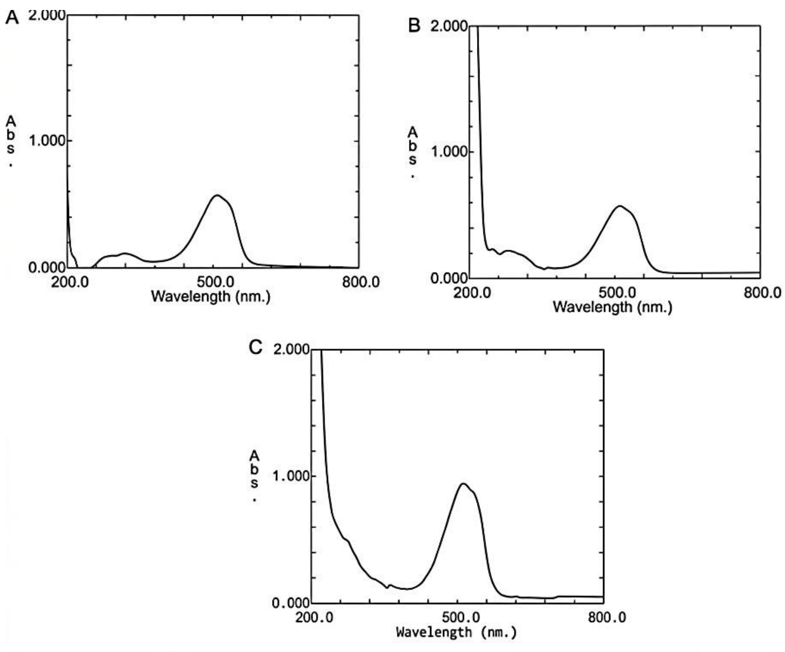

Figure 2. UV–Visible spectra of free methyl orange and immobilized methyl orange in the HYPR-modified silica matrix.

To further investigate the pH sensitivity of the immobilized indicator, the monoliths were exposed to alkaline medium and their spectral response was monitored over time. As shown in Figure 3, a gradual decrease in the absorption band associated with the acidic form of methyl orange was accompanied by the appearance of a new absorption band corresponding to its basic form.

The observed spectral shift reflects the conversion of methyl orange from its protonated state to its deprotonated state and confirms that the immobilized indicator retained its pH-responsive characteristics after encapsulation. These findings demonstrate that the immobilized methyl orange remains chemically active and capable of responding to environmental pH variations.

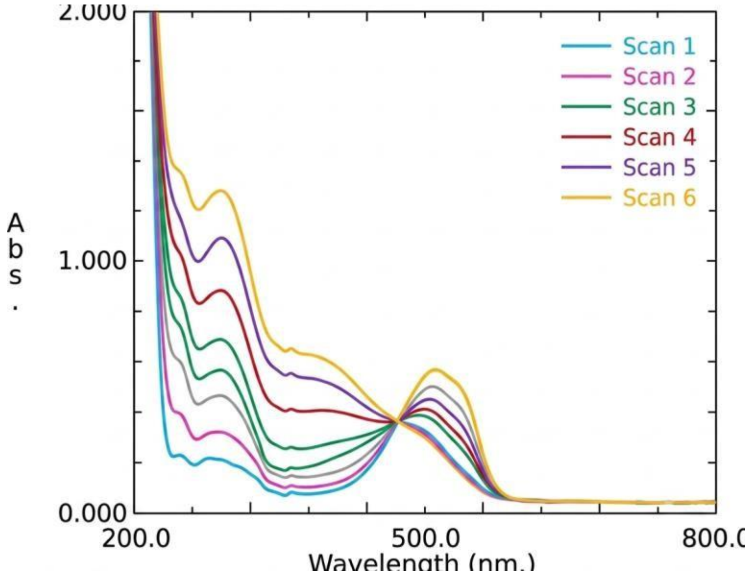

Figure 3. UV–Visible spectra of immobilized methyl orange after exposure to alkaline medium at different time intervals.

3.4. Surface Morphology

The surface morphology of the prepared silica monoliths was investigated using White Light Interferometry (WLI). Representative three-dimensional surface images are presented in Figure 4.

The HYPR-modified silica matrix exhibited a porous and interconnected microstructure characteristic of sol–gel-derived materials. The presence of the surfactant influenced the organization of the silica framework and promoted the development of a more open network structure suitable for incorporation of indicator molecules.

The morphology of the methyl orange-loaded monoliths differed slightly from that of the blank silica matrix, indicating successful incorporation of dye molecules within the silica framework. Surface roughness analysis revealed a heterogeneous microstructure associated with the distribution of methyl orange throughout the porous network.

The observed surface characteristics support the successful immobilization of methyl orange and demonstrate the role of the HYPR surfactant in modifying the internal architecture of the silica matrix.

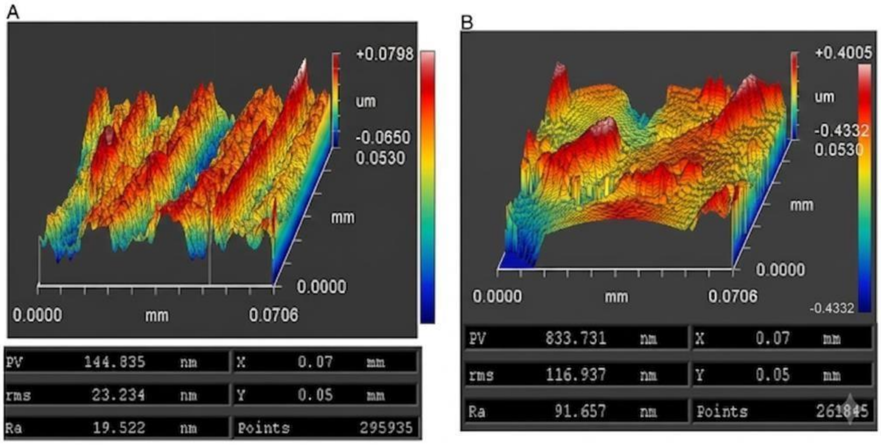

Figure 4. White Light Interferometry images of (a) HYPR-modified silica matrix and (b) methyl orange immobilized in the HYPR-modified silica matrix.

3.5. pH Response of Immobilized Methyl Orange

The pH response of the immobilized methyl orange was evaluated by exposing the prepared monoliths to acidic and alkaline solutions. A distinct color transition was observed, corresponding to the characteristic behavior of methyl orange in solution.

Under acidic conditions, the immobilized indicator exhibited the color associated with its protonated form, whereas exposure to alkaline media produced the characteristic color of the deprotonated form. The spectral changes observed in Figure 3 further confirm this transformation.

These results demonstrate that the immobilization process preserved the functionality of methyl orange and allowed the indicator to maintain its sensitivity toward changes in pH. Consequently, the HYPR-modified silica matrix represents a promising platform for the development of solid-state optical pH sensors.

4. Conclusion

Methyl orange was successfully encapsulated within a HYPR-modified silica matrix using an acid-catalyzed sol–gel process. The prepared monoliths exhibited good transparency, structural stability, and effective dye immobilization. UV–Visible spectroscopy confirmed that the optical properties and pH-responsive behavior of methyl orange were preserved after encapsulation, while surface morphology analysis demonstrated successful incorporation of the indicator within the porous silica framework.

The HYPR-modified silica matrix provided a suitable environment for dye immobilization and facilitated interaction between the encapsulated indicator and the surrounding medium. The results indicate that the developed hybrid material has considerable potential for application as a solid-state pH sensing platform and may serve as a basis for future optical sensing technologies.

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