**Tuning of Catalytic Property Controlled by the Molecular Dimension of Palladium–Schiff Base Complexes Encapsulated in Zeolite Y**

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**ABSTRACT:** Planar palladium–Schiff base complexes are synthesized, maintaining the order of their molecular dimensions as PdL1 < PdL2 < PdL3 < PdL4 < PdL5 in free state, as well as encapsulated in zeolite Y, where L1: N,N′-bis(salicylidene)ethylenediamine and L2, L3, L4, and L5 are derivatives of L1. All encapsulated complexes have shown better catalytic activity for the sulfoxidation of methyl phenyl sulfide in comparison to their homogeneous counter parts. These hybrid systems are characterized with the help of different characterization techniques such as X-ray diffraction analysis, scanning electron microscopy—energy-dispersive X-ray spectrometry, X-ray photoelectron spectroscopy, Fourier transform infrared, and UV–visible spectroscopy; all of these studies have suggested that the largest complex deviates by the maximum from its free-state properties, and a radical change in the reactivity of the complex is observed.

**INTRODUCTION**

The selective oxidation of thioethers is a significant reaction in organic synthesis because the product sulfoxides are the important intermediates of various biological advances in natural compounds. These products also have versatile applications in agrochemical, pharmaceutical, and polymer industries and also can be used as ligands in asymmetric catalysis, oxo-transfer reagents, and solvents. They often play an important role as therapeutic agents such as antiulcer, antibacterial, antifungal, antiatherosclerotic, antihelmintic, antihypertensive, and vasodilators. As a result, there are substantial amounts of work done to develop varieties of competent catalysts for the sulfoxidation process. Most of the conventional synthesis routes for the sulfoxidation process are quite efficient, but they require the use of lots of organic and inorganic oxidants and solvents which obviously leads to the production of a large amount of toxic waste. Consequently, it is essential to develop a “greener approach” to synthesize the desired sulfoxidation products. To adopt the environmentally benign methodology, there should be some modification in the traditional procedures such as uses of environment-friendly oxidants, solvent-free reaction medium, and eco-friendly catalysts. Aqueous hydrogen peroxide is a most popular oxidant for the sulfoxidation process because of its easy handling, greater availability, and eco-friendly approach for the reaction due to the formation of water as the only byproduct. Schiff base transition-metal complexes have played a vital role in the progress of catalytic processes with greater effectiveness in biological and industrial applications. However, the homogeneous catalysts generally have some disadvantages in the catalytic processes such as their instability, difficulty in the separation, lack of reusability, and lack of greener approach.

The modern catalytic science favors those catalysts, which can conquer the limitation of these homogeneous catalysts without the loss of reactivity. In this direction, heterogenization of conventional homogeneous catalysts is a suitable route to couple the reactivity of the complex with the additional shape and size selectivity and site isolation properties appended by the host materials. Encapsulation of transition-metal complexes in different hosts such as microporous and mesoporous materials is an alternative contemporary approach to accomplish the synthesis of designer catalysts. To synthesize designer catalysts, especially biomimetic systems, zeolites are found as competent hosts for the encapsulation of transition-metal complexes having a molecular dimension comparable with that of the cavity of the host zeolites.

Among those, employment of zeolite-encapsulated metal complexes in the field of oxidation or epoxidation of alkanes, alkenes, alcohols, and aromatic compounds has been extensively studied and reported, but comparatively fewer reports are available on the sulfoxidation process. Maurya and co-workers have investigated different vanadium complexes entrapped in zeolite Y and explored their catalytic activities for the oxidation and sulfoxidation reactions. The authors have stated that the encapsulated complexes in zeolite Y have shown almost a similar reactivity for the sulfoxidation reaction when compared with their free states. However, higher turnover frequencies and reusability make the zeolite-encapsulated complexes more suitable than their free-state analogues.

**Supporting Information**
In the present work, we have synthesized some novel palladium–Schiff base complexes in the encapsulated state in zeolite Y as well as in the free state. The molecular dimensions of the complexes follow the order as PdL1 < PdL2 < PdL3 < PdL4 < PdL5 (where L1: N,N′-bis(salicylidene)-ethylenediamine, L2: N,N′-bis(5-hydroxy-salicylidene)-ethylenediamine, L3: N,N′-bis(5-bromo-salicylidene)-ethylenediamine, L4: N,N′-bis(5-methyl-salicylidene)-ethylenediamine, and L5: N,N′-bis(5-methoxy-salicylidene)ethylenediamine) (given in Figure 1).

These systems are employed as catalysts for the sulfoxidation of methyl phenyl sulfide to study the steric and electronic effect of different substituent groups which are attached on the complexes and their consequences on the reactivity upon encapsulation. These systems are characterized by powder X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), IR, and UV–visible (UV−vis) spectroscopy. It is quite fascinating to observe that as compared to their neat forms, encapsulated complexes have shown quite a significate control over the reactivity of the encapsulated systems.

### RESULTS AND DISCUSSION

**Elemental Analysis.** The pure Na-zeolite Y has the unit cell formula as Na_{89}Si_{130}O_{388}·2H_{2}O, and the Si/Al ratio is 2.34. The elemental analysis (EDXS) spectra of the encapsulated complexes PdL1, PdL2, PdL3, PdL4, and PdL5 has suggested that the Si/Al ratio remains nearly unaffected even after the encapsulation for all cases (shown in Table 1), indicating that no significant dealumination takes place during the process of encapsulation. The wt % data of palladium metal ions in the Pd-exchanged zeolite and encapsulated metal complexes (given in Table 1) highlight the reasonable observation that the concentration of palladium metal ions in Pd-exchanged zeolite Y is greater than those of all zeolite-entrapped complexes. It is perhaps obvious because in the process of encapsulation, slight leaching of metal ions may occur which eventually leads to the reduction of the wt % of palladium in encapsulated metal complexes.

**Scanning Electron Microscopy.** Scanning electron microscopic studies of pure zeolite Y and the Soxhlet extracted hybrid PdL1-Y, PdL2-Y, PdL3-Y, PdL4-Y, and PdL5-Y systems are carried out (the SEM images are shown in Figure 2). Zeolite boundaries are evidently observable in the SEM images of zeolite-entrapped complexes and also comparable with that of the pure zeolite Y which is a clear manifestation of the absence of unreacted species or impurities on the surface of the host lattice in the final host–guest products.

**Powder XRD Analysis.** Powder XRD patterns of the parent zeolite Y, Pd-zeolite Y, and palladium−salen complexes encapsulated in zeolite Y have been recorded (shown in Figure 3). The complexes are synthesized within the supercage of zeolite Y via the flexible ligand synthesis method, and the unreacted ligands and complexes are completely removed by the Soxhlet extraction technique; however, during the whole process, the host framework is remained intact. The X-ray diffractograms of PdL1-Y, PdL2-Y, PdL3-Y, PdL4-Y, and PdL5-Y samples and that of the pure zeolite Y exhibit no shift in peak positions when compared. This observation is indeed a significant one as it specifies the preservation of host lattice integrity even after the encapsulation of a large complex into it. However, XRD patterns of PdL1-Y, PdL2-Y, PdL3-Y, PdL4-Y, and PdL5-Y evidently illustrate a substantial reversal in the intensity of the peaks at 2θ = 10° and 12°, that is, I_{2θ} < I_{111}. In comparison to those in the XRD patterns of pure and Pd-exchanged zeolite Y. This intensity reversal has already been empirically correlated with the existence of the large complex.
inside the cavity of zeolite Y.21 Interestingly, a tethered complex on the host surface does not show such type of intensity reversal in XRD patterns. Moreover, the absence of new peaks in the XRD patterns of palladium-encapsulated complexes is a signature of the formation of the complexes in low concentrations inside the host lattice.

X-ray Photoelectron Spectroscopy. XPS is an important technique which provides an indirect proof about the complex formation in free as well as encapsulated states. All core constituent elements of the complexes such as C (1s), N (1s), O (1s), and Pd (3d) of PdL1, PdL1-Y, PdL2-Y, and PdLS-Y complexes are presented in the form of survey spectra and high-resolution spectra (Figures 4 and 5 and Figures S1–S3 in the Supporting Information), and the corresponding binding energy data are tabulated in Table 2. The low concentration of palladium in the encapsulated complexes makes the XPS signal for metal weak which is actually in accordance with the concentration-dependent EDXS, IR, and UV–vis spectroscopic studies. The observed data evidently indicate the presence of C, N, O, Na, Si, Al, and Pd in their respective chemical states, which is in accordance with the literature.26d,22 Intense and broad carbon (1s) XPS spectra have been observed for palladium–salen complexes in both the states, are further deconvoluted into two peaks, and confirmed the presence of sp3 and sp2 carbon atoms in those complexes. Similarly, these complexes have shown the XPS signals for nitrogen (1s), oxygen (1s), and validated for the (M–N), (N=C) and (C–O), (M–O) elemental state, respectively.22e All encapsulated complexes show a very weak signal for palladium metal because of its low loading level within the host lattice. High-resolution XPS spectra of palladium in the PdL1 complex have shown two signals at the binding energies of 335.96 and 340.96 eV which are attributed to 3d5/2 and 3d3/2, respectively. However, when the complex is encapsulated in zeolite Y, 3d5/2 and 3d3/2 XPS signals are broader and appear at the almost same binding energies (335.13 and 340.49 eV) and additionally a new signal originates at a relatively higher binding energy of 347.87 eV. This additional XPS signal at a higher binding energy has been observed for all encapsulated complexes (PdL1-Y, PdL2-Y, and PdLS-Y) but not for the free-state complex (PdL1). The higher shift in binding energy is definitely a consequence of the removal of electron density from the metal center.16d,22 In this context, it could have relevant significance as the geometry which the metal complex adopts under encapsulation might play the major role in the depletion of electron density from the metal center. Furthermore, XPS signals of Na (1s), Si (2p), and Al (2p) elements are also observed at their expected positions in the XPS spectra of all encapsulated complexes.

IR Spectroscopy. Fourier transform infrared (FTIR) spectral data of ligands and palladium–Schiff base complexes in neat as well as encapsulated states along with the pure zeolite Y have been recorded (Figure 6 and Table S1 in the Supporting Information). IR spectroscopic data have provided information about the retention of integrity of the zeolite Y framework and also indicated the successful complex formation within the supercage of zeolite Y. The four major zeolitic IR bands are observed in the region of 450–1200 cm–1, and two additional peaks appear at 1643 and 3500 cm–1. The IR bands at 560, 717, 786, and 1018 cm–1 are attributed to (Si/Al–O)4 bending mode, double ring, symmetric stretching, and asymmetric stretching vibrations, respectively,23 whereas IR bands at 3500 and 1643 cm–1 are assigned to surface hydroxylic group and lattice water molecules, respectively.20a All of these bands mostly remain unaffected for all of the encapsulated systems (PdL1-Y, PdL2-Y, PdL3-Y, PdL4-Y, and PdL5-Y) because the host framework does not experience any structural modifications during the process of encapsulation. However, the salen ligands and corresponding complexes are primarily identified by the IR studies as C==N stretching, C==C stretching, and C–O stretching vibrations of the salen ligands appear at the expected positions. Upon complexation, these FTIR bands show an essentially identical vibration with shifts to lower energies as an
outcome of the coordination with the metal ion. IR peaks of the encapsulated complexes are mostly difficult to identify because of the appearance of the strong zeolitic bands in the 450–3500 cm$^{-1}$ region. Advantageously, some characteristic IR peaks of

Figure 4. High-resolution XPS signals of (A) Pd (3d), (B) C (1s), (C) N (1s), and (D) O (1s) for PdL1 complex (black-colored graphs are experimental data, and green- and blue-colored peaks are peak-fitted data).

Figure 5. High-resolution XPS signals of (A) Pd (3d), (B) C (1s), (C) N (1s), and (D) O (1s) for PdL1-Y complex (black-colored graphs are experimental data, and green- and blue-colored peaks are peak-fitted data).
relatively low intensities are observed in the range of 1600−1200 cm\(^{-1}\) which is mainly due to the guest complex and are suitable to study because zeolitic IR bands are silent particularly in this region. These bands in encapsulated complexes have appeared with little shifts with respect to that of the nearly planar free-state complex. These shifts could be attributed to the altered geometry the complex adopts to accommodate itself in the rigid host supercage under encapsulation. Furthermore, a shift in C−H deformation bands has already been identified as an indication of the encapsulation of the complex inside the zeolite Y.\(^{24}\)

**Solid-State UV−Vis Spectroscopy.** Electronic spectra of the ligand and corresponding complex in solution (Figure S4 in the Supporting Information) or solid states have been studied for the justification of complex formation. In addition to that, comparative optical spectra of palladium−Schiff base complexes in both states have been studied thoroughly to understand the nature of the geometry they adopt under encapsulation (given in Figure 7 and Table 3). Electronic transitions of the Schiff

| s. no. | samples | Si (2p) | Al (2p) | C (1s) | N (1s) | O (1s) | Pd (3d\(_{5/2}\)) | Pd (3d\(_{3/2}\)) |
|--------|---------|---------|---------|--------|--------|--------|----------------|----------------|
| 1      | PdL1    | 284.04, 282.82 | 285.23, 283.65 | 399.11, 396.54 | 532.84 | 335.92 | 340.96 |
| 2      | PdL1-Y  | 101.52, 75.15 | 285.23, 283.65 | 398.88, 397.72 | 530.96 | 335.13 | 340.49, 347.87 |
| 3      | PdL2-Y  | 101.55, 72.94, 73.79 | 285.23, 283.65 | 398.81, 397.76 | 530.95 | 335.39 | 340.21, 346.72 |
| 4      | PdL5-Y  | 102.72, 73.16, 75.47 | 285.66, 283.64 | 399.93, 397.88 | 532.63, 530.69 | 337.11 | 343.34, 346.64 |

Figure 6. FTIR spectra of encapsulated palladium−salen complexes in zeolite Y: (a) pure zeolite Y, (b) PdL1-Y, (c) PdL2-Y, (d) PdL3-Y, (e) PdL4-Y, and (f) PdL5-Y.

Figure 7. (A) UV−vis spectra of (a) PdL1 and (b) PdL1-Y; (B) UV−vis spectra of (a) PdL2 and (b) PdL2-Y; (C) UV−vis spectra of (a) PdL3 and (b) PdL3-Y; (D) UV−vis spectra of (a) PdL4 and (b) PdL4-Y; and (E) UV−vis spectra of (a) PdL5 and (b) PdL5-Y.
base ligands appearing in the range of 210–240 nm are assigned to the $\pi-\pi^*$ transition and in the range of 240–300 nm are assigned to the $n-\pi^*$ transition. However, in the free-state transition-metal complexes, these transitions are shifted relatively toward a lower energy; $\pi-\pi^*$ transition has been observed in the range of 210–240 nm, and $n-\pi^*$ transition has been observed in the range of 240–300 nm. The most evident confirmation of free-state complex formation is the appearance of charge-transfer (CT) and d–d transitions bands, which are clearly observed in the solid-state electronic spectra in the range of 335–450 nm for different palladium–Schiff base complexes. However, the appearance of d–d bands in square planar palladium complexes is indistinguishable in comparison with those of nickel–salen complexes. In palladium–salen complexes, these bands appear in the range of 335–450 nm, whereas in nickel complexes, these are identified in the 510–570 nm region.26 According to the ligand field theory, for 4d series elements, d orbital splitting is much higher as compared to that of the corresponding 3d series elements and hence d–d bands in palladium–salen complexes have shifted toward the high-energy region and merged with CT bands. After encapsulation, palladium–salen complexes have shown a comparable pattern; however, the CT and d–d bands which are instigated from the metal center are primarily affected, that is, intensified26 and blue-shifted on encapsulation.24 Such an observation eventually signifies the deviation of the free-state geometry of the complexes when encapsulated, especially around the metal center. Interestingly, PdL2-Y, PdL3-Y, PdL4-Y, and PdL5-Y complexes have shown a blue shift in greater extent compared to that observed for PdL1-Y because these complexes have larger molecular dimensions and might experience more space constraint imposed by the topology of the supercage which in turn causes more distortion in the geometry of these complexes.

**Catalytic Study.** Transition-metal complexes are efficient catalysts for sulfoxidation reaction using aqueous hydrogen peroxide as an oxidant.27 However, among the zeolite-encapsulated transition-metal complexes, palladium–Schiff base complexes are comparatively less investigated catalysts for sulfoxidation. Therefore, in the present report, free-state as well as encapsulated Pd complexes have been employed as catalysts for sulfoxidation reaction in the presence of $\text{H}_2\text{O}_2$: the objective is to, however, investigate how encapsulation affects the catalytic activity of the complex. For all cases, methyl phenyl sulﬁde is converted to corresponding sulfoxide selectively (Scheme 3), and it is quite interesting to note that the catalytic activities of these encapsulated complexes in terms of percentage conversion as well as turnover number (TON) are significantly higher than those of their corresponding free state (catalytic data are given in Table 4).

**Table 4. Oxidation of Methyl Phenyl Sulﬁde after 4 h Reaction Time with $\text{H}_2\text{O}_2$ as an Oxidant**

| s. no. | samples | % conversion | TON | selectivity for sulfoxide | selectivity for sulfone |
|-------|---------|--------------|-----|--------------------------|-------------------------|
| 1     | pure zeolite Y | 7 | 82.1 | 17.9 |
| 2     | PdL1 | 39 | 30 | 98.66 | 1.33 |
| 3     | PdL1-Y | 48 | 201 | 97.25 | 2.74 |
| 4     | PdL2 | 30 | 25 | 99.51 | 0.49 |
| 5     | PdL2-Y | 60 | 477 | 98.85 | 1.04 |
| 6     | PdL3 | 63 | 67 | 98.98 | 1.01 |
| 7     | PdL3-Y | 71 | 564 | 95.45 | 4.54 |
| 8     | PdL4 | 36 | 30 | 98.31 | 1.68 |
| 9     | PdL4-Y | 76 | 623 | 92.01 | 7.98 |
| 10    | PdL5 | 26 | 23 | 98.18 | 1.81 |
| 11    | PdL5-Y | 86 | 835 | 92.23 | 7.76 |

Surface impurities in the form of ligands and surface complexes are minimized by extensive Soxhlet extraction using different solvents, and the uncoordinated palladium ions are removed by further ion-exchange reaction with sodium ions. The exercise of removal of impurities is truly significant so as to comprehend better the origin of modified catalysis with respect to the parent zeolite Y as it has been observed that the parent zeolite Y (Na-zeolite Y) shows fairly less % conversion as well as selectivity (shown in Table 4). Though another direction of research could be with the change in Si/Al ratio as it definitely affects the catalytic activity of the encapsulated complexes,28 the present work highlights on the catalysis of hybrid host–guest systems with the Si/Al ratio of the host kept constant by taking the same lot of parent zeolite Y for the synthesis of all encapsulated complexes.

The reactivity order of free-state complexes follows the order as PdL3 > PdL1 ≈ PdL4 > PdL2 > PdL5, whereas after encapsulation, the order of reactivity is just in accordance with the molecular dimension of the complexes and it is PdL5 > PdL4 > PdL3 > PdL2 > PdL1. The proposed mechanism for this catalytic reaction includes the nucleophilic attack of $\text{H}_2\text{O}_2$ on the electropositive metal center. As a result, in the transition state, electron-rich nucleophile is bonded through the axial position to the metal complex. Plausible mechanism for the oxidation of sulfides to the corresponding sulfoxides catalyzed by the PdL complexes entrapped in zeolite Y using $\text{H}_2\text{O}_2$ as an oxidant is given in Scheme 1, and it is obvious that the electropositive character of the metal drives the first step of the reaction.29

Bhadbhade and Srinivas have already reported that an electron-withdrawing group ($\text{Cl}^-$) makes the copper–salen complex distorted from its square planar geometry,30 whereas an electron-donating group (–OCH$_3$) on the same position tries to maintain the planarity of the complex, especially around the metal. Distortion around the metal atom by any means makes the complex more susceptible to nucleophilic attack by removing electron density from the metal center; on the contrary, the metal center in planar effectively conjugated systems becomes electron-rich and less efficient for the nucleophilic attacks. We also have observed a similar effect...
for the nickel−salen complexes encapsulated in zeolite with different molecular dimensions in series.\textsuperscript{25}

Currently, while exploring the catalytic activity of the Pd complexes in their free states, PdL3 having electron-withdrawing (−Br) groups is identified as the most efficient catalyst which is just in line with the previous arguments. Electron-withdrawing (−Br) groups in PdL3 make the complex distorted even in the free state and subsequently more reactive. Electron-releasing groups (−OH and −OCH\textsubscript{3} in PdL2 and PdL5, respectively) make those palladium−salen complexes less reactive. PdL1 and even PdL4 complexes showing nearly the same TON have exhibited intermediate reactivity, as they do not have a noteworthy push−pull effect of substituents. As expected, the PdL5 complex has shown most exciting modified enhancement in reactivity on encapsulation, as the complex is most distorted to be fitted into the supercage because of its largest molecular dimensions. The modified reactivity of the encapsulated complexes is indeed a consequence of the geometry they adopt under encapsulation in zeolite Y. It is quite evident and well-supported by the optical spectroscopic and XPS studies and even theoretical studies\textsuperscript{25} that the rigid spherical walls of the zeolite supercage impose significant space constraint which in turn leads the complexes to suffer from noteworthy structural distortion. The observed relative shifts toward higher values of binding energy in XPS signals for the zeolite-encapsulated complexes indicate the enhancement of the electropositive character of the metal center after encapsulation. Distortion around the metal atom by any means makes the complex more susceptible to nucleophilic attack by removing electron density from the metal center; on the contrary, the metal center in planar effectively conjugated systems becomes electron-rich and less efficient for the nucleophilic attacks.

The reusability of PdL5-Y is evaluated for the same sulfoxidation reaction (shown in Figure 8); the catalyst could be effectively used for up to four cycles without much loss in catalytic activity. In conclusion, it can be stated that the reactivity of free-state complexes is mainly governed by the electronic factor, whereas after encapsulation, the catalytic activity order of the complexes is in accordance with the molecular dimensions or the extent of distortion.

\section*{CONCLUSIONS}

Zeolite-encapsulated complexes are indeed better heterogeneous catalysts with enhanced adapted reactivity for the oxidation process, for example, sulfoxidation reaction. The comparative studies of palladium−Schiff base complexes in free as well as encapsulated states provide clear insights about the modified reactivity for the catalytic oxidation processes of such systems after encapsulation. The observed blue shift in d−d bands in the electronic spectra of encapsulated complexes has demonstrated the effect of the space restrictions imposed by rigid host frameworks on the coordination environment around the metal as space restrictions compel the encapsulated guest complex to adopt an unusual nonplanar geometry for better accommodation inside the supercage to minimize the van der Waals interaction. High-resolution XPS spectra of Pd (3d) of encapsulated complexes showing an additional signal toward a higher binding energy also confirms the change in the electronic environment around the metal upon encapsulation. Comparative catalytic studies of these hybrid systems provide a fascinating correlation between modified structural aspects and adapted functionality of complexes, and therefore it can be concluded as the degree of distortion in the structure of the encapsulated complex is the key point for the remarkable modified catalytic activity of the systems.

\section*{EXPERIMENTAL SECTION}

\textbf{Materials and Preparation.} Pure zeolite Y is purchased from Sigma-Aldrich, India. Salicylaldehyde and its derivatives and ethylenediamine are purchased from Alfa Aesar, and Sigma-Aldrich, India. Salicylaldehyde and its derivatives and ethylenediamine are purchased from Sigma-Aldrich, India. All solvents (ethanol, acetone, methanol, and diethyl ether) are purchased from S.D. Fine, India.

\textbf{Preparation of Ligands (L1, L2, L3, L4, and L5).}\textsuperscript{24,31} Two molar ratios of salicylaldehyde (or its derivatives) dissolved in ethanol is refluxed for 10−15 min. One molar ratio of ethylenediamine is added into it. The reaction mixture is refluxed for 30 min at 60−70 °C and then ice-cooled for an hour. Bright yellow solid flakes are obtained as a product, thoroughly washed with ethanol, and then dried in air (given in Scheme 2).

\textbf{Preparation of Complexes.}\textsuperscript{32} Ligands (L1, L2, L3, L4, and L5 in the respective reactions) taken in ethanol are refluxed, and then aqueous solution of equimolar ratio of palladium acetate is added dropwise into them. The reaction
mass is further refluxed for 30 min. For the synthesis of these Schiff base complexes, an inert environment is used. The final greenish-yellow colored product is recovered, washed with ethanol and diethyl ether, and then dried at room temperature (Scheme 2).

Preparation of Palladium-Exchanged Zeolite Y. Pure Na-zeolite Y (10 g) (Na58Al58Si136O388·yH2O) is allowed to disperse in 0.01 M palladium salt [Pd(CH3COO)2 = 0.224 g] in 100 mL of water to acquire the required loading level of palladium ions and stirred at room temperature for 24 h. The slurry is filtered, washed repeatedly with water, and then desiccated for 12 h at 150 °C (Scheme 2).

Synthesis of Encapsulated Pd(II)−Schiff Base Complexes in Zeolite Y.24b,31 Schiff base ligands are flexible in nature; thus, one of the methods of encapsulation of metal complexes inside the supercage of zeolite Y could be possible via the “flexible ligand” approach (Scheme 2). The palladium-exchanged zeolite and excess amount of the ligand (L1, L2, L3, L4, and L5 in each respective reaction) are allowed to react at 200−250 °C for 24 h under constant stirring to synthesize the complex inside the supercage of zeolite Y. The reaction mass is then recovered and further subjected to the Soxhlet extraction with the different solvents such as acetone, methanol, and diethyl ether in a sequence. The product is dried in a muffle furnace for 10−12 h at 150 °C. The recovered material is further reacted with 0.01 M NaCl solution for 12 h to remove the unreacted metal ions, followed by filtration and washing until the filtrate is negative for the chloride ion test.

Sulfoxidation of Methyl Phenyl Sulfoxide. Aqueous 30% H2O2 (0.57 g, 5 mmol), methyl phenyl sulfoxide (0.62 g, 5 mmol), and the catalyst (0.015 g) are mixed in a minimum amount of solvent (3 mL of CH3CN), and the reaction mixture is stirred at 60 °C temperature for 4 h. The progress of the reaction is monitored by gas chromatography (GC) at different time intervals, and the products are identified and quantified (by using an internal standard method) with the help of GC (Scheme 3). The calibration curve of thioanisole is provided in the Supporting Information (Figure S5). To attain the maximum efficiency, reaction conditions are optimized by varying the different reaction parameters such as temperature, time duration of reaction, and amount of catalysts, considering the PdL1-Y complex as the representative catalyst (Figure S6 is given in the Supporting Information).
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