

Article

MNP (M = Zn, Cu, and Ag) Catalyst Embedded onto Zeolite Y Surface for Efficient Dye Reduction and Antimicrobial Activity

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Abstract: This paper deals with synthesizing Zn, Cu, and AgNPs supported on the surface of zeolite Y for catalytic and antimicrobial applications. Firstly, the zeolite Na-Y was exchanged with solutions containing metal precursors and then a chemical treatment was used to transform the metal cations into metal nanoparticles. The different samples were characterized by different characterization methods. The reduction of methylene blue (MB) and orange (OG) dyes in the presence of NaBH₄ and nanocatalysts in a simple and binary system showed good results. It was shown in this study that the concentration of the reagents, the nature of metal species, and the nature of the dye can influence the conversion of the dye. The calculated k_{app} obtained by the best catalyst (Ag/Y) in a simple system was 1.882 min^{−1} and 1.115 min^{−1} for MB and OG dyes, respectively. It was found that the Ag/Y catalyst was more selective via MB in the binary system containing OG+MB dyes. The reuse of the Ag/Y catalyst in five cycles showed good results via the conversion of the MB dye without losing its performance. For antimicrobial activities, encouraging results have been recorded on different strains having inhibition zones between 14 and 25 mm.

Keywords: zeolite Y; metallic nanoparticles; dye reduction; antimicrobial activity



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1. Introduction

The development of technology in the industrial sector has caused a significant problem, particularly in the pollution of air, soil, and water sources [1–4]. The textile industry is classified among the most harmful sources of pollution following the numerous releases of dyes. In general, the textile industries use stable dyes, which makes the waste challenging to process. It has been found that slight concentrations of dyes can create serious illnesses that can lead to death due to their carcinogenic nature [5,6].

To date, several treatment processes have been used to limit this problem. Among the most used methods, there are oxidation reactions [7,8], photocatalysis [9,10], hydrogenation [11–15], adsorption [16,17], separation [18,19], coagulation–flocculation [20,21], and other biological treatments [22,23]. The scientific community has shown particular interest in finding efficient and rapid processes for the treatment of wastewater. The reduction or hydrogenation of dyes is one of the reactions that can meet these requirements following the speed of the process, and the obtained products can be used in other applications, particularly in organic synthesis [24–27]. In addition, this treatment process can significantly reduce the toxicity of water polluted by dyes or transition metals [28–30]. The work of Yu et al. showed that the reduction process can be applied even for toxic transition metals like Cr(VI), which can be reduced to Cr(III) [29].

In general, without a catalyst or reducing agent, the reduction of dyes cannot be achieved. In the work of Hachemaoui et al., they found that the reduction of dyes under the presence of NaBH_4 alone does not lead to any transformation [31,32]. Meanwhile, the addition of a few milligrams of catalyst causes a rapid reaction [31,32]. This clarifies that the function of the catalyst is to move electrons from the electron source (NaBH_4) to the receptor (dye) [33]. Among the keys that can influence the kinetics of the reaction is the choice of nanoparticles, which can play a crucial role in the transfer of electrons as well as in the speed of the reaction, which is the objective of this work [31,34].

According to a review of the literature, metallic nanoparticles have a high performance in transferring electrons, which makes them good candidates for this type of reaction [35–38]. However, the presence of nanoparticles in the solution leads to a known problem in their tendency to form aggregates, which reduces their application in the catalysis field. Among the most used attempts to prevent this problem is to stabilize them in organic or inorganic matrices. To prevent the aggregation of nanoparticles, several supports have been proposed, including clays, polymers, activated carbon, mesoporous silica, and zeolites. Each solid requires a special treatment to stabilize the nanoparticles, which is why most solids use the impregnation method. However, it has been confirmed that large particles can be formed when using this method. Ion exchange on zeolites is considered to be one of the most promising methods for localizing nanoparticles in specific sites, so ultrafine nanoparticles can be obtained that do not exceed the size of the zeolite nanopores. In addition, the nanoparticles loaded onto the zeolite surface can be well stabilized in the pores as a result of the multiple interactions between the nanoparticles and the zeolite framework, which limits their leaching into the reaction medium. It should also be noted that the nature of the zeolites and their pores has a significant influence on the content and size of the nanoparticles. Zeolite Y was chosen in this work because of its unique adsorption and surface properties, which are different from those of other zeolites.

In this study, zeolite Y was used due to its remarkable surface properties, its pore openings, and also its specific surface area, which exceeds $800 \text{ m}^2/\text{g}$ [39]. Zeolite Y is a hydrated zeolite with a cubic structure (space group $\text{Fd-}3\text{m}$). It has a super cage whose entrance diameter can reach $7.4 \text{ \AA}$. Ion exchange is among the most remarkable properties of these zeolites. In fact, Na^+ neutralizes the negative charges coming from aluminum during its isomorphic substitution by silicon [40]. The choice of a different metal cation was to select the best nanoparticles with high catalytic performance via the reduction in organic pollutants. To this end, the selectivity of zeolite Y via the retention of transition metals was tested using an ion exchange process between the Na^+ -containing zeolite Y and the cations Zn, Cu, and Ag. The exchanged materials were subsequently chemically treated with a reducing agent to be used as catalysts via the reduction of MB and OG dyes in a simple, binary system. It should also be noted that this method of catalyst preparation allows for

the production of ultrafine, well-dispersed nanoparticles compared to other non-porous or macroporous materials.

In this work, zeolite Y was used to confine different metal nanoparticles (M = Zn, Cu, and Ag) using cation exchange followed by treatment with NaBH₄ solution. The properties of these catalysts will be tested via the reduction of the MB and OG dyes in the simple and binary system, and according to the optimized conditions, a reaction mechanism will be proposed. It is known that metal species based on Zn, Cu, and Ag have antimicrobial properties, which is why we have chosen to test our supported materials via antibacterial and antifungal applications.

2. Results

2.1. XRD Analysis

The XRD patterns of the faujasite samples before and after modification by the metal nanoparticles (MNPs) (M: Ag, Cu, and Zn) are shown in Figure 1. As shown in the XRD patterns of sample Y, all the diffraction peaks characteristic of faujasite-type zeolite are present [37]. These peaks are intense and fine, and no additional peaks were observed. This proves that the material is pure and well crystallized. For the Cu/Y and Zn/Y samples, the XRD patterns only indicate the presence of a faujasite-type structure (Figure 1). The absence of peaks corresponding to the diffraction of copper and zinc nanoparticles can be justified by the existence of these metals in low quantities on the one hand (see EDX results) and their strong dispersion on the surface of the zeolite Y somewhere else. The XRD patterns of the Ag/Y sample show, in addition to the faujasite phase, the presence of four diffraction peaks at $2\theta = 38.2^\circ$, 44.3° , 64° , and 77° corresponding to the plane (111), (200), (220), (311) characteristics of the face-centered cubic (c.f.c) structure of metallic silver AgNPs (JCPDS-04-0783). Thus, we can confirm that AgNP nanoparticles are well confined in our Ag/Y material [41,42]. It should be noted that the decrease in the intensity of the diffraction lines of faujasite is not due to a loss of crystallinity but to a strong absorption of AgNPs in X-rays [43]. To confirm the presence of Zn, Cu, and Ag NPs on the surface of zeolite Y, XPS analysis was performed (see Figure S1). As shown in this figure, all characteristic peaks of zeolite Y (O1s, Si2p, and Al2p peaks) are present. According to the high-resolution XPS spectra of Zn2p, Cu2p, and Ag3d, it is clear that the metal cations were reduced after NaBH₄ modification.

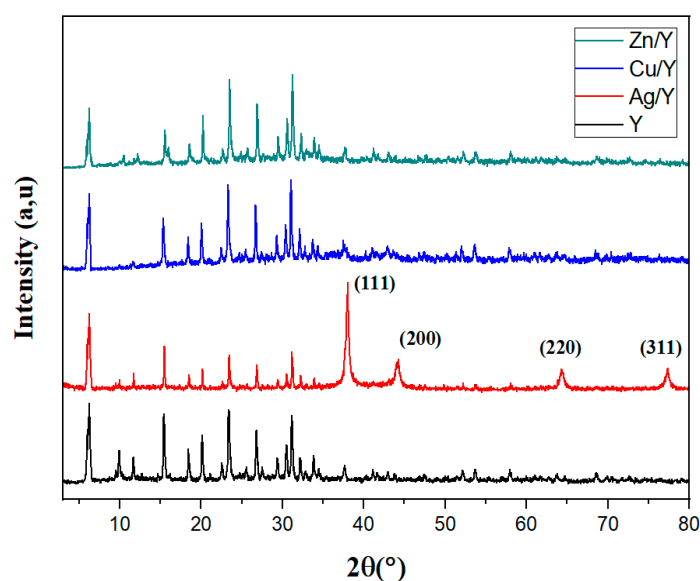


Figure 1. XRD patterns of zeolite Y, Zn/Y, Cu/Y, and Ag/Y.

2.2. FTIR

The FTIR spectra of the Ag/Y, Cu/Y, and Zn/Y nanocatalysts show the presence of all the vibration bands of the starting zeolite Y (Figure S2). This finding suggests that the structure of zeolite Y was maintained upon modification by metallic MNPS. Among the main bands observed, an intense band is located between 980 and 1062 cm^{-1} attributed to the symmetric and asymmetric stretching vibrations of TO_4 (T: Si, Al) [11]. The band at 3400 cm^{-1} is attributed to the vibration bands of Si-OH and OH hydroxyl groups [11], and a weak band at 1646–1626 cm^{-1} is attributed to the OH group originating from the adsorption of H_2O [44]. The weak band located at 500–550 cm^{-1} is linked to the secondary building units (SBUs) of the zeolite [44], while the band located between 450 and 500 cm^{-1} corresponds to Metal-O vibrations [45].

2.3. UV-Vis Reflectance Spectroscopy

UV-vis spectroscopy was used to determine the coordination state of Ag, Cu, and Zn species confined in faujasite. As shown in Figure S3, the zeolite containing the Zn species has a band between 250 and 300 nm, which corresponds to a Zn/ZnO mixture. The band centered at 220 nm was attributed to the existence of Zn^{2+} cations, thus indicating a partial reduction of zinc [17]. The copper-based sample was characterized by a broad band at 450–580 nm assigned to the absorption of Cu(0) nanoparticles and another at 340 nm corresponding to Cu_2O species [46]. The band located at 300–380 nm indicates the presence of zero-charge Ag nanoparticles [47], which is in agreement with the XRD results.

2.4. SEM/TEM

The morphology of a catalyst plays a crucial role in its activity. For this reason, all the samples obtained were analyzed by SEM. The SEM images of the modified samples (Cu/Y, Ag/Y, and Zn/Y) are shown in Figure 2. The morphology of the different nanocatalysts is similar to the morphology of zeolite Y [40]. Their size is between 0.4 and 0.7 μm . As shown in this figure, the areas circled in yellow show the existence of ultrafine particles corresponding to MNP nanoparticles (M: Zn, Cu, and Ag) [43,48]. Moreover, the existence of these elements in zeolite Y was also confirmed by EDX analysis (see Figure S4). All the constituents of these catalysts were present (Si, Al, Ag, Cu, Zn, Na, and O). As indicated in this figure, the percentage of nanoparticles (Ag, Cu, and Zn) differs from one catalyst to another; hence, the Ag/Y catalyst presented the highest content of AgNPs on its surface, and this confirms the results obtained by XRD. The increase in the NPS content in zeolite Y was accompanied by a decrease in the sodium content. This result indicates that the dispersion of the metal nanoparticles in the zeolite Y followed an ionic exchange process between Na^+ and metal cations (before chemical treatment with NaBH_4). As shown in Figure 3, the metal nanoparticles are well formed on the surface of zeolite Y. The Zn/Y sample clearly shows large sizes of nanoparticles due to their aggregation. However, the Cu/Y and Ag/Y samples presented ultrafine particles that were well dispersed on the surface of the Y zeolite. The size of nanoparticles varies from 25 to 15 nm for these two samples.

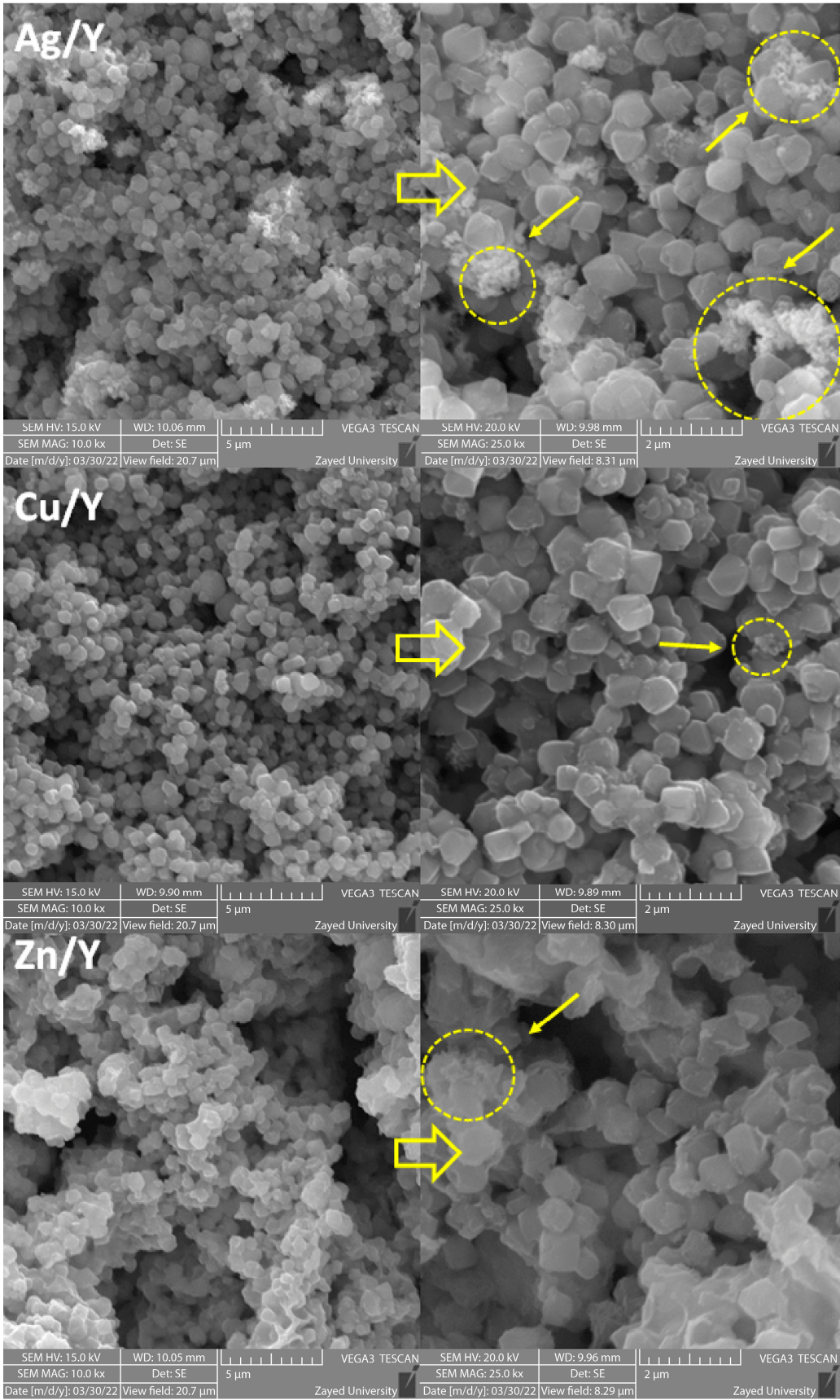


Figure 2. SEM images of obtained catalysts.

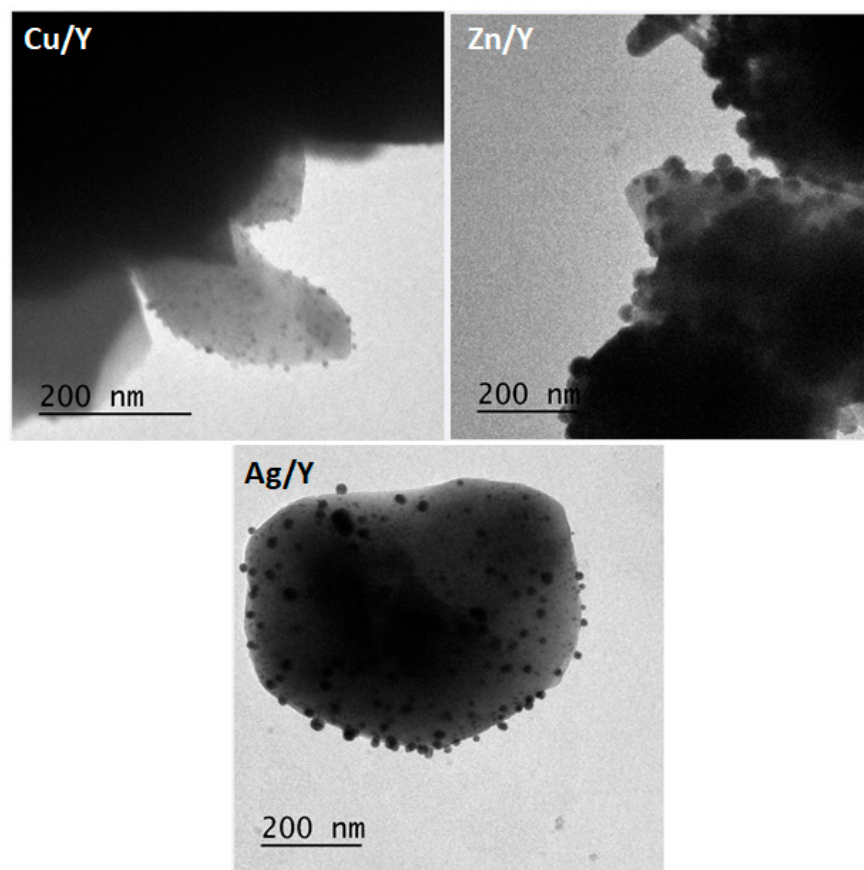


Figure 3. TEM images of Cu/Y, Zn/Y, and Ag/Y.

3. Catalytic Application of MNPs/Y

The catalytic reduction of dyes by NaBH_4 is a practical degradation method that is not only feasible in terms of efficiency and cost but also environmentally friendly as it provides end products that are less toxic and useful for other applications, particularly in organic synthesis [40]. In order to optimize the best reaction conditions, catalytic hydrogenation of the obtained materials was tested in the degradation of MB and OG dyes.

3.1. Effect of Nanocatalyst Nature

The metallic species supported on the Y zeolite structure can powerfully affect their catalytic properties [31,43,49]. For this, the different catalysts were used toward MB dye reduction using only 3 mg of each catalyst (Ag/Y, Cu/Y, or Zn/Y), 2 mL of an MB solution, and 1.5 mL of freshly prepared NaBH_4 (5 mM).

The evolution of the UV-Vis spectra of MB dye in the presence of Cu/Y, Zn/Y, and Ag/Y catalysts shows that the absorption band at 664 nm characteristic of the MB dye decreases rapidly when using Cu/Y and Ag/Y catalysts (Figure 4). This band completely disappears after 8 min of reaction for Cu/Y and 4 min for Ag/Y, indicating a total MB conversion to Leuco-MB (see Figure 4a–d). The ineffectiveness of the catalyst Zn/Y is due to the low content of this metal in the zeolite Y (see the EDX analysis) and also to a partial reduction of the Zn^{2+} cations, which is in agreement with the results found previously. The catalytic performance of different catalysts varies in the following order: Ag/Y > Cu/Y > Zn/Y.

Based on the results, the catalyst Ag/Y was the most efficient due to the high content of AgNPs confined to the surface of the zeolite Y. The calculated k_{app} for the different catalysts (see Figure 4e) is as follows: 1.882 min^{-1} , 0.4909 min^{-1} , and 0.0062 min^{-1} for Ag/Y, Cu/Y, and Zn/Y, respectively.

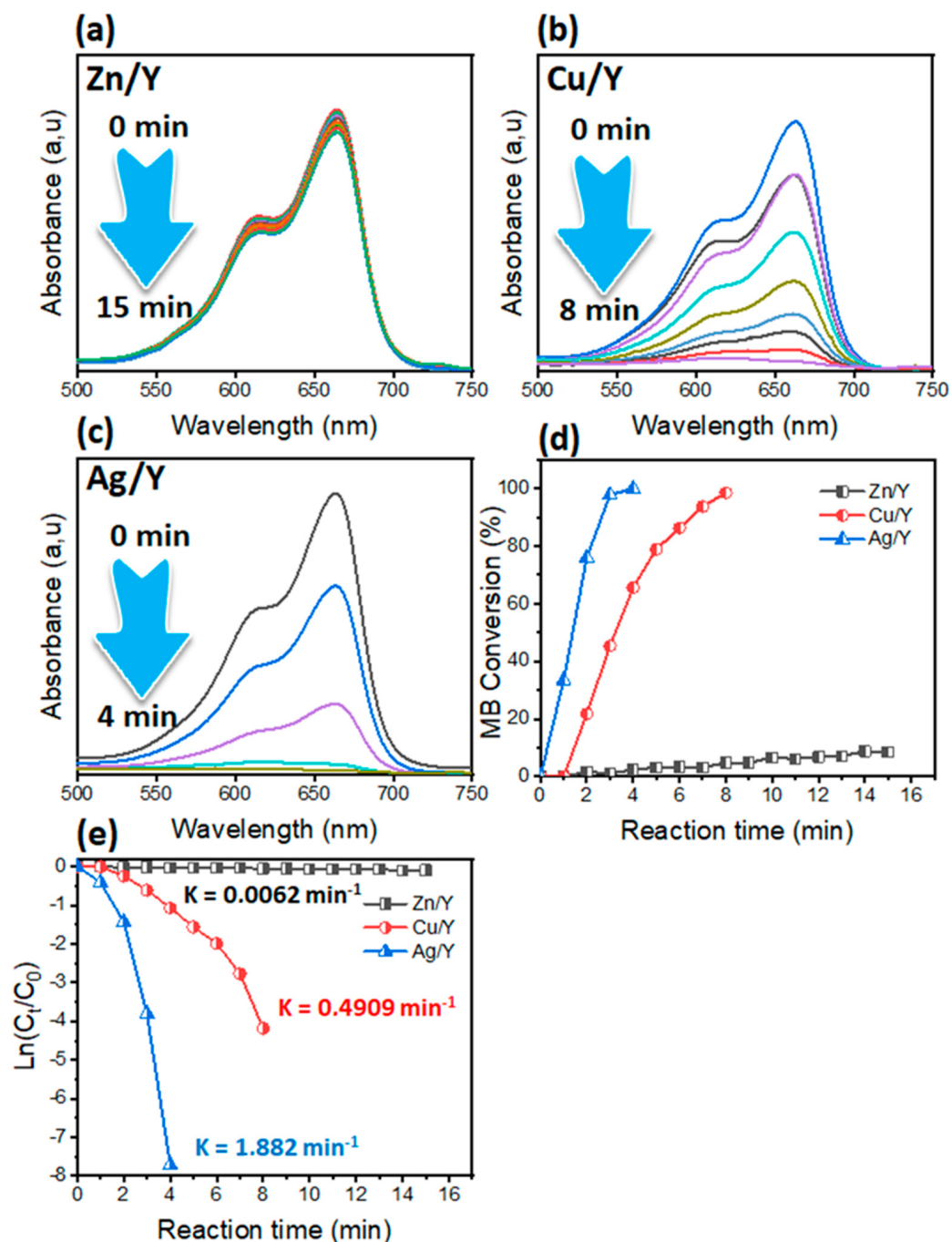


Figure 4. UV-vis of MB dye during reduction in the presence of catalysts: (a) Zn/Y, (b) Cu/Y, and (c) Ag/Y. (d) MB conversion versus time. (e) Plot of $\ln(C_t/C_0)$ versus time.

The rate constant calculated for the best catalyst Ag/Y is more critical than the values obtained in Table 1. This catalyst works well because it can remove organic species even at lower NaBH₄ concentrations and more significant concentrations of organic contaminants ([NaBH₄] = 5 mM, and [MB] = 0.6 mM). The Ag/Y material was used as a catalyst for the rest of the catalytic tests.

Table 1. Comparative study.

Dyes	Catalyst	Catalyst Mass (mg)	[NaBH ₄] (mM)	Reaction Time (min)	K _{app} (min ⁻¹)	Ref.
MB	AgNPS-MCM-41	3	6.8	7	0.29	[50]
	Cu@SBA-3	3	6.8	3	2.944	[32]
	Cu@SBA-3	3	6.8	3	2.944	[32]
	12.5Cu@SBA-15	1	20	8	0.51	[51]
	Fe ₃ O ₄ @C-TiO ₂ -Ag	10	/	10	0.38	[52]
	Cu(4%)-ALG(ZnNPs)	5.1	1.6	09	0.798	[53]
OG	Ag/Y	3	5	4	1.882	This study
	NFCP	2	20	4	0.4514	[54]
	NFSP	2	20	4	0.4227	[54]
	Cu(4%)-ALG(ZnNPs)	5.1	1.6	19	0.252	[53]
	Ag/Y	3	5	6	1.115	This study

3.2. Effect of Catalyst Masse

In this study, the effect of catalyst mass was investigated by varying the mass of Ag/Y from 1 to 3 mg. The dye concentrations for MB and NaBH₄ were 0.6 mM and 5 mM, respectively. As shown in Figure 5, the time required for the disappearance of the MB dye increased as the catalyst mass decreased. A time of 4 min was sufficient for the complete conversion of MB dye to Leuco-MB when only 3 mg of Ag/Y catalyst was used. According to the literature, the decrease in the mass of the catalyst leads to a reduction in the silver nanoparticles and, consequently, to a slowing down of the conversion of MB to Leuco-MB [31]. The $k_{app}/\text{catalyst mass}$ ratio (calculated using 3 mg Ag/Y) was $0.627 \text{ min}^{-1} \cdot \text{mg}^{-1}$, which is a significant value compared to previously published works (see Table 1). This also indicates the ability of this catalyst to reduce organic pollutants even at low masses.

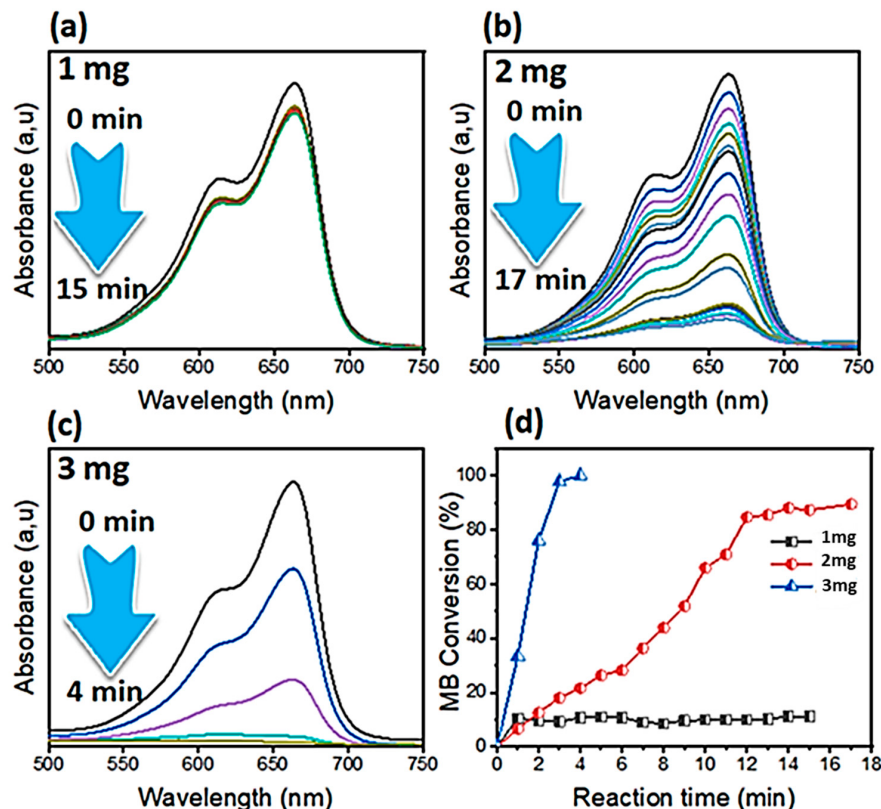


Figure 5. UV-vis of MB dye during reduction in the presence of different masses of catalyst Ag/Y: (a) $m = 1$ mg, (b) $m = 2$ mg, and (c) $m = 3$ mg. (d) MB conversion versus time.

3.3. Effect of Reagent Concentration

The nature and kinetics of the reaction can be affected by the concentration of the reactants. For this, we studied these parameters by varying the MB dye and NaBH_4 concentration. Figure 6a,b demonstrate that the Ag/Y catalyst reduced the MB dye more quickly at a lower MB concentration (0.3 mM) than at a higher MB concentration (0.6 mM). This tendency ensures that there are enough sites even at low MB concentrations. Therefore, the ratio of MB molecules to sites increases when MB concentrations increase, which lengthens the reaction time [32,55].

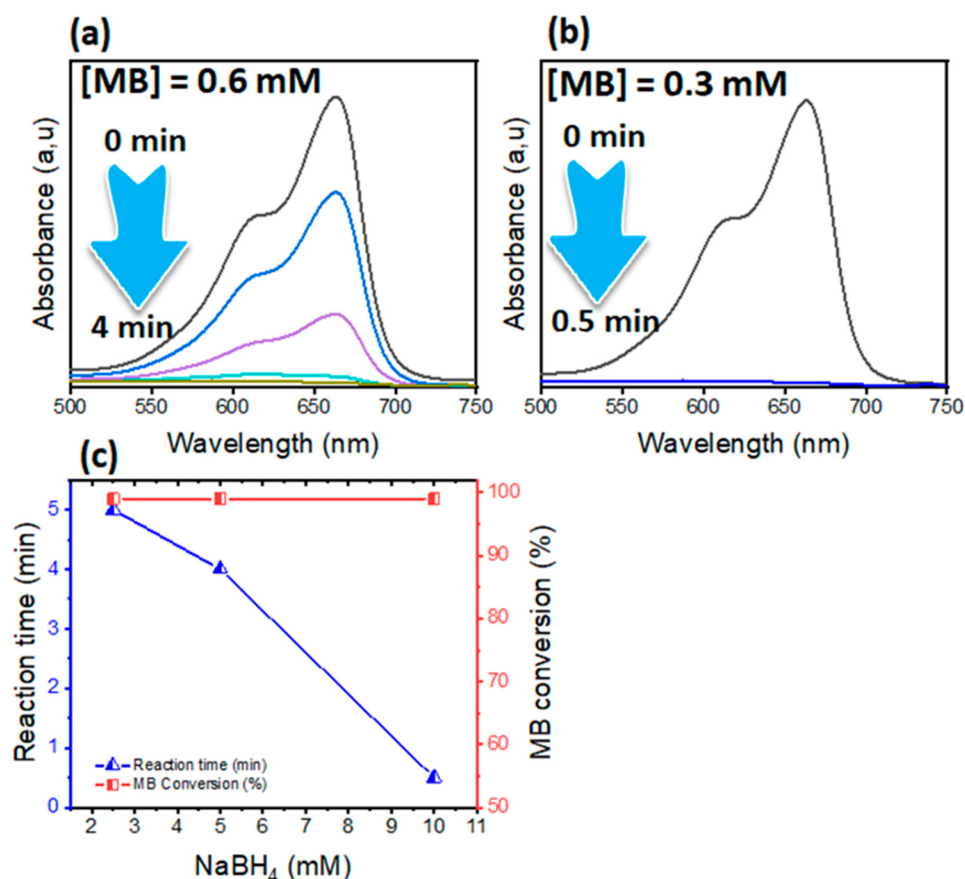


Figure 6. UV-vis of MB dye during reduction in the presence of different concentrations of MB: (a) [MB] = 0.6 mM; (b) [MB] = 0.3 mM. (c) Correlation curve between initial NaBH_4 concentration, reaction time, and MB conversion.

Figure 6c shows the variation in reaction time MB conversion as a function of NaBH_4 concentration. It is evident that the concentration of NaBH_4 has a direct effect on the MB dye conversion and reaction time. When the concentration of NaBH_4 was increased, more MB dye was converted in a shorter reaction time. Therefore, as the concentration of NaBH_4 decreases, the reaction time increases. The role of NaBH_4 in this study is that it is a reducing agent that can transfer electrons that the nanoparticles then transfer to the MB dye [33,55,56]. As the concentration of NaBH_4 increases, the electron density increases, resulting in an easy and rapid reduction of the MB dye to Leuco-MB. The effect of NaBH_4 without a catalyst does not result in any product, even at higher concentrations of NaBH_4 . This also shows the synergy between the nanoparticles and NaBH_4 in donating and transporting electrons to the acceptor MB dye.

3.4. Reduction of Azo Dye (OG)

In this part, the best conditions optimized previously in a reduction of OG dye were studied using the Ag/Y catalyst. We provide a general overview of the conversion time of OG and its differences from that of the MB dye (Figure 7a–c). According to the same conditions, the reduction of the OG dye is a little slow (6 min) compared to MB (4 min); this is related to the catalyst's surface nature, which is reflected by a significant induction area. It should be noted that the OG dye is an anionic molecule, which makes its diffusion slow in a matrix containing silver nanoparticles coated with electrons provided by NaBH_4 . The calculated value of the rate constant was 1.115 min^{-1} greater compared to the materials presented in Table 1. According to these results, our prepared Ag/Y material showed its effectiveness via two different types of pollutants, which can broaden its field of application, particularly for the purification of polluted water.

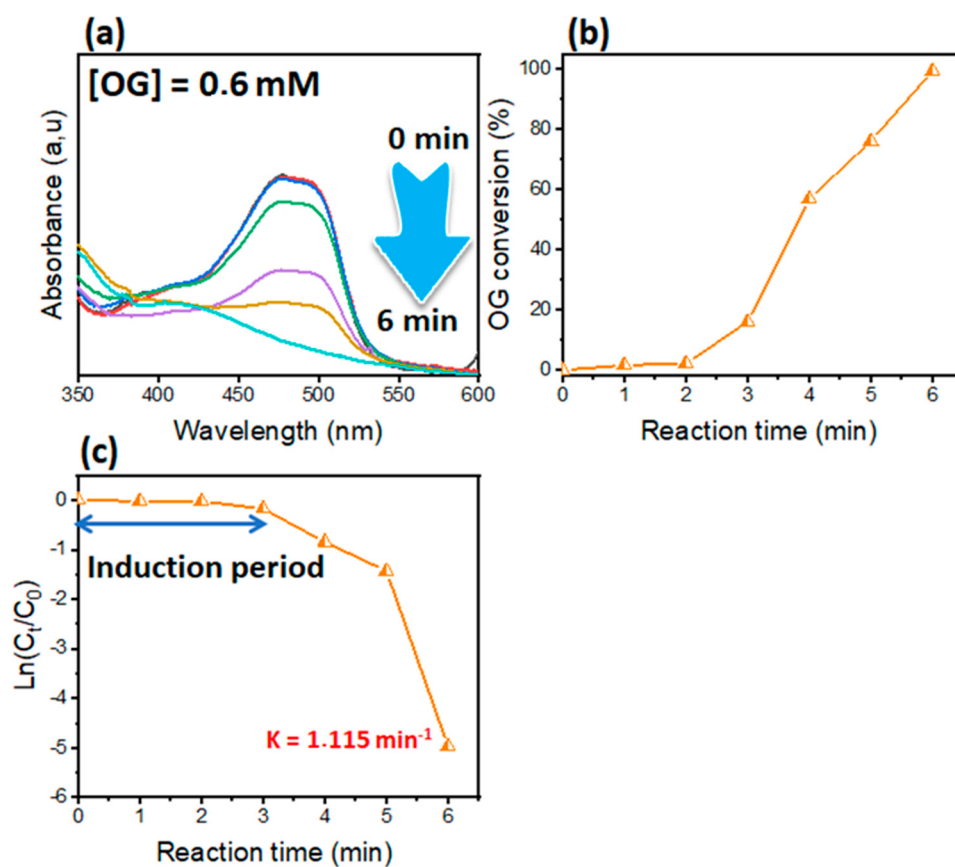


Figure 7. (a) UV-vis of OG dye during reduction in the presence of catalyst Ag/Y. (b) OG conversion versus time. (c) Plot of $\ln(C_t/C_0)$ versus time.

3.5. Reduction of MB and OG in a Binary System

The mixture between MB and OG dyes was studied to test the behavior and selectivity of the catalyst Ag/Y. As shown in Figure 8, the MB and OG dyes have preserved their characteristic bands, confirming that their interactions are almost negligible. It was confirmed that the bands of MB and OG dyes decreased over time, which confirms the performance of this catalyst in reducing dyes even in the mixture [43,56,57]. The catalyst Ag/Y was found to be more selective with MB dye; therefore, the reaction time to convert MB to Leuco-MB took only 1 min. Therefore, the hydrogenation of the OG dye took 3 min to achieve 99% conversion. Notably, the reaction time was shorter than in the simple system; nevertheless, this was caused by the reaction medium containing a low dye concentration. According to the literature, the strong interactions between the cationic MB dye and the electronic layer

on the surface of silver metallic nanoparticles produced from the dissociation of NaBH_4 can account for the catalyst Ag/Y's selectivity toward the MB dye [43,56,57]. The efficacy of this catalyst is confirmed by its capacity to reduce the dye mixture in optimal conditions, which include a high dye concentration and a low concentration of NaBH_4 (5 mM). These findings are highly relevant to the previous investigations [43,56,57].

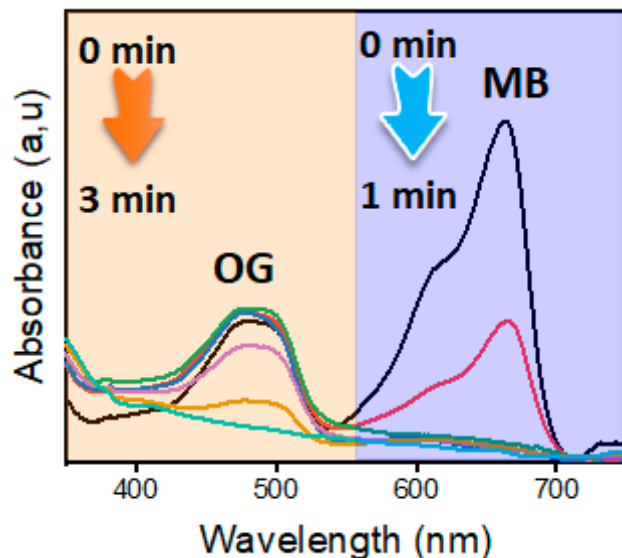


Figure 8. UV-vis of a mixture of dyes (MB + OG) during reduction in the presence of catalyst Ag/Y.

3.6. Reuse of Catalyst Ag/Y

The reuse of catalyst Ag/Y was studied in order to test its performance and stability in the reaction medium. The reduction of MB dye was selected as a model reaction, and the catalyst was just washed with distilled water after each cycle before being reused. The leaching of Ag species was verified by the precipitation test with NaCl and by XRF analysis, the results of which confirmed that the catalyst was more stable during the reuse tests. As shown in Figure 9, the catalyst Ag/Y was more efficient during the different cycles studied, but a slight increase in the reaction time was observed, probably due to the fixation of the reagents on its surface, which is in agreement with the literature [56–58].

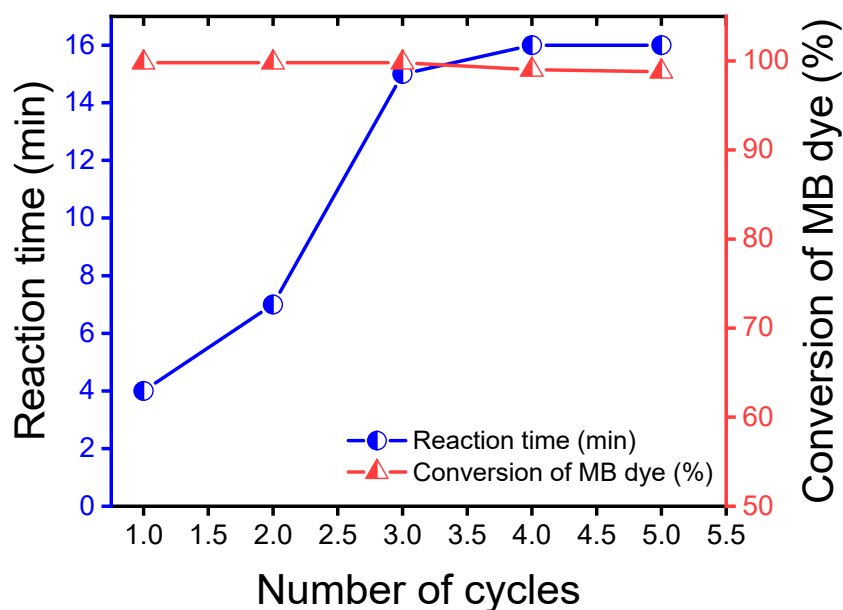


Figure 9. Reuse of catalyst Ag/Y.

3.7. Reaction Mechanism

The hydrogenation mechanism of the two dyes can include several steps (see Figure 10). First, the reagents diffuse onto the surface of nanocatalysts (AgNPs). According to the results obtained, it was shown that the two dyes do not undergo the same path following their diffusions, which are different. According to the results obtained, it was noted that the diffusion of OG toward the sites goes through an induction period; then, for the case of the dye MB, its diffusion was faster following its cationic form, which is more favorable to be attached to the surface of AgNPs, which are charged by electronic layers and hydrides resulting from the dissociation of NaBH_4 . The exact role of AgNPs is to transfer electrons from NaBH_4 to the acceptor (MB or OG).

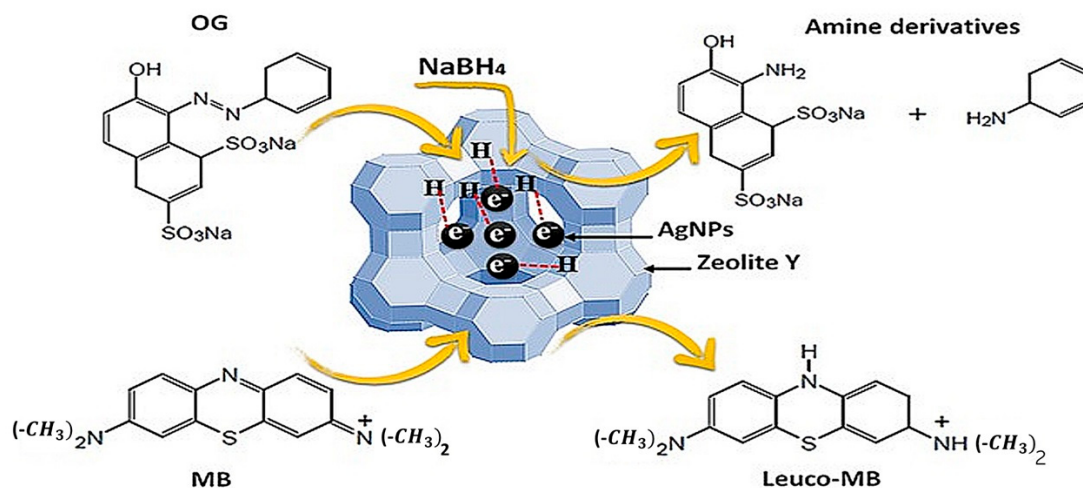


Figure 10. Mechanism for reduction of MB and OG dyes on catalyst Ag/Y.

In the case of the MB dye, it undergoes a single hydrogenation step at the $\text{C}=\text{N}$ -bond level to $\text{CH}-\text{NH}$ -. The final product, Leuco-MB, is characterized by its transparent color [59,60]. From an analysis point of view, it is effortless to analyze it by UV-vis spectroscopy. Hence, forming a new band located at 258 nm confirms the formation of Leuco-MB (see Figure S6). While the OG dye undergoes two consecutive stages of hydrogenation, the first stage of hydrogenation leads to the formation of hydrazine in the azo group ($-\text{N}=\text{N}-$). This product is unstable and will subsequently undergo a second hydrogenation, subsequently causing the breakdown of the hydrazine and leading to the formation of amine derivatives, which are characterized by the appearance of the new bands at 260, 316, and 351 nm [61–63] (see Figure S6).

4. Antifungal and Antibacterial Activities

The results of the antibacterial and antifungal activities of materials Cu/Y, Zn/Y, and Ag/Y on Gram-positive (*Staphylococcus aureus* ATCC 25923, 43300) and Gram-negative (*Escherichia coli* ATCC 25922) bacterial species and on the fungal species (*Pseudomonas aeruginosa* CIP A22) are illustrated in Table 2 and Figure 11. The three composites presented good antibacterial and antifungal activity, which is justified by the presence of a clear halo around the disks of the M/Y composites. The degree of sensitivity of the *Staphylococcus aureus* ATCC 25923 bacteria to the three composites is almost equivalent. However, the Cu/Y composite showed a significant inhibitory effect toward the bacterium *Staphylococcus aureus* ATCC 43300. A strong sensitivity toward the Zn/Y composite of the bacterium *Escherichia coli* ATCC 25922 was also observed. The diameter of the inhibition zone is approximately 23 mm. The variation in the antibacterial activity of different samples can be attributed to the size of the nanoparticles and their dispersion on the surface of zeolite Y.

According to previously published studies, the small size of the nanoparticles facilitates their penetration into the bacteria's membrane. This structural change in the membrane causes cellular degradation of the bacteria, which can lead to its death [64].

Table 2. Results of biological tests.

Bacteria	Zn/Y	Cu/Y	Ag/Y
<i>Escherichia coli</i> ATCC 25922	23	15	16
<i>Staphylococcus aureus</i> ATCC 25923	21	20	19
<i>Staphylococcus aureus</i> ATCC 43300	19	25	16
<i>Pseudomonas aeruginosa</i> A22	15	14	25

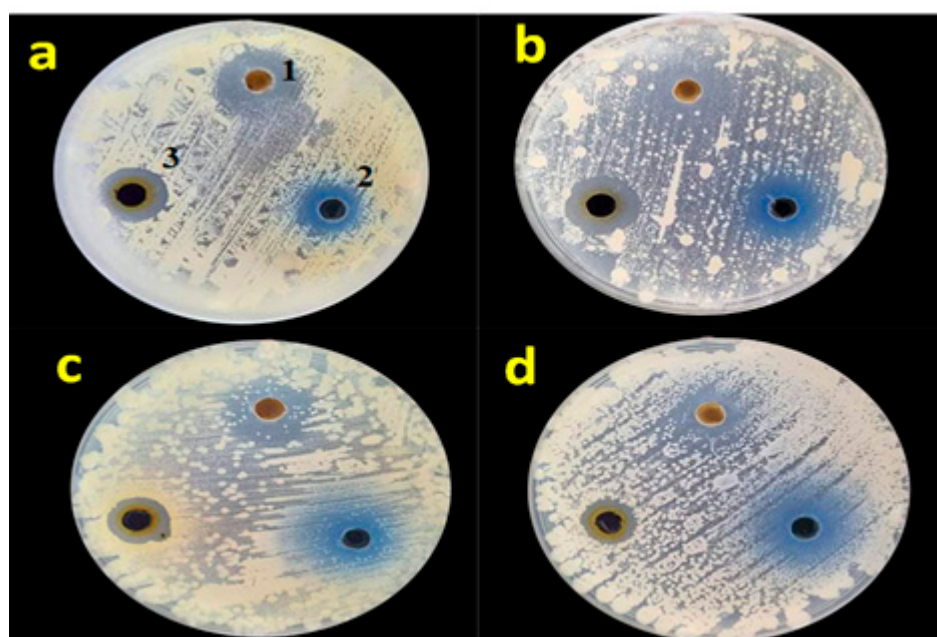


Figure 11. Antibacterial and antifungal activities of Zn/Y (1), Cu/Y (2), Ag/Y (3). (a) *Escherichia coli* ATCC 25922; (b) *Staphylococcus aureus* ATCC 25923; (c) *Staphylococcus aureus* ATCC 43300; (d) *Pseudomonas aeruginosa* A22.

The fungal strain *Pseudomonas aeruginosa* A22 showed high sensitivity toward the Ag/Y material with an estimated inhibition zone diameter of approximately 25 mm. This result is in agreement with the bibliographic data, which indicate the good antifungal activity of Ag nanoparticles against fungal pathogens [62–64].

5. Experimental

5.1. Chemicals and Reagents

The reagents used in this study include the following: Ludox HS (40% SiO₂ 60% H₂O, Sigma-Aldrich, Saint Louis, MO, USA), sodium aluminate NaAlO₂ (50.38% Al₂O₃ 36.70% Na₂O 13.63% H₂O, Sigma-Aldrich), sodium hydroxide in pellets NaOH (99%, Prolabo, Tokyo, Japan), zinc diacetate (Zn(OAc)₂ Sigma-Aldrich), copper nitrate (Cu(SO₄)₅ 5H₂O, Sigma-Aldrich), silver nitrate (Ag(NO₃), Merck, Boston, MA, USA), sodium borohydride (NaBH₄, 98%, Sigma-Aldrich), methylene blue (MB, Sigma-Aldrich), orange G (OG, Sigma-Aldrich), and demineralized water.

5.2. Preparation of Zeolite Y

The Na-Y zeolite gel with a composition 5 Na₂O, 10 SiO₂, Al₂O₃, and 160 H₂O was prepared from an alkaline solution containing sodium hydroxide and sodium aluminate. After 1 h of stirring, the silica was added dropwise. The mixture thus formed was homogenized for 2 h at room temperature and then brought to its crystallization temperature at 100 °C for a period of 48 h. The obtained solid was washed with demineralized water and dried for 24 h at 80 °C.

5.3. Preparation of MNPs/Zeolite Y

Zeolite Y was modified with different transition metals (M: Ag, Cu, and Zn) in order to test their performance via dye reduction and antimicrobial activities. For this, the Ag, Cu, and ZnNPs were prepared as follows. Approximately 100 mL of a solution 0.1 M containing metal precursors (M: Zn, Cu, or Ag) was put in a beaker, and then 1 g of zeolite Y was added and stirred for 2 h. The obtained solids are washed with demineralized water, filtered, and dried at 60 °C. The exchanged metal cations Cu(II), Zn(II), and Ag(I) were converted into metal nanoparticles by treating them with 100 mL of a NaBH₄ solution following 1 h of stirring. The obtained materials were collected by filtration, washed with deionized water, and dried at 60 °C for 24 h. The characterization methods used for determining the properties of different samples are given in the Supplementary Materials.

5.4. Hydrogenation Reaction of MB Dye

The reduction of the dye (MB) catalyzed by the different obtained materials and in the presence of NaBH₄ was carried out according to a protocol described in the literature [33,43]. For this, several parameters were studied, namely the effect of the nature of the metallic species supported on zeolite Y, the effect of the dose of the catalyst, and the effect of the concentration of reagents. The reduction was performed as follows: 2 mL of the dye at a known concentration was poured into a quartz basin and then a quantity of catalyst was added. The suspension obtained after the addition of 1.5 mL of NaBH₄ was subjected to UV-vis (UV-vis Specord 210, Analytik, Jena, Germany) and then analyzed. The MB dye conversion and rate constant were calculated by Equations (1) and (2), respectively.

$$\text{Dye conversion (\%)} = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

$$\ln \frac{C_t}{C_0} = -K_{app} \times t \quad (2)$$

where

k_{app} (s^{−1}): is the rate constant;

C_0 (mM): is the initial dye concentration;

C_t (mM): is the final dye concentration.

5.5. Hydrogenation of a Mixture of Dyes MB + OG

The simultaneous reduction of the two dyes MB+OG was carried out under the following conditions: 3 mg of Ag/Y catalyst, the concentration of MB and OG dyes in the mixture is 0.15 mM, and the concentration of NaBH₄ is equal to 5 mM. The volume of the dye mixture is 2 mL and 1.5 mL of NaBH₄. The reaction mixture is placed in a cuvette and subsequently transferred to UV-vis.

5.6. Methylene Blue (MB) Adsorption Test

To find out if the disappearance of MB dye is due to its adsorption on the surface of the catalyst or to the reduction of MB to Leuco-MB, we added 3 mg of the catalyst Ag/Y in

contact with 2 mL of a solution of MB dye with a concentration of 0.3 mM and 1.5 mL of demineralized water in the absence of NaBH₄. The adsorption test was followed by UV-vis (Figure S5).

6. Conclusions

Zeolite Y with high crystallinity and high purity was synthesized hydrothermally and then modified with silver, copper, and zinc metallic nanoparticles. Characterization by X-ray diffraction proved the dispersion of the NPs inside the Y zeolite without loss of crystallinity. The presence of the NPs in a metallic state (zero charge) was confirmed through UV-vis analysis. The use of the produced materials in the reduction of methylene blue (MB) shows that the kind of metal scattered on the zeolite Y affects the conversion of methylene blue into leuco-methylene blue. The catalytic performance of different samples increases in the following order: Ag/Y > Cu/Y > Zn/Y. The reduction time of MB was considerably lowered by increasing the concentration of NaBH₄. The simultaneous reduction of two dyes, MB (cationic) and OG (anionic), showed the selectivity of the reduction reaction via the MB dye. The Ag/Y catalyst was successfully used for three successive cycles without performance loss, confirming its effectiveness and stability. The antibacterial and antifungal tests carried out on the bacterial strains *Escherichia coli* and *Staphylococcus aureus* and the fungal strain *Pseudomonas aeruginosa* A22 made it possible to highlight the bactericidal and fungicidal power of the M/Y catalyst through the formation of inhibition zones.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/catal15050407/s1>, Figure S1: FTIR spectrum of nanocatalysts M/Y; Figure S2: UV-visible spectrum of zeolite Y modified by MNPs (M: Ag, Zn, and Cu); Figure S3: EDX spectra of Ag/Y, Cu/Y, and Zn/Y samples; Figure S4: UV-vis of MB and OG dyes before and after the catalytic test: (a) MB dye; (b) OG dye. Figure S5. Adsorption test of MB by Ag/Y. Figure S6. XPS spectra a: Ag/Y; b: Cu/Y; c: Zn/Y.

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