



PHOTOCATALYTIC DEGRADATION OF SOME DYES USING La/Ti/ZSM-5

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ABSTRACT:

Modified photocatalytic materials were prepared by incorporation of lanthanum ions on TiO₂ supported by ZSM-5. These materials were characterized by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), UV-Vis diffuse reflectance spectra (UV-Vis DRS), and surface area (*BET*) measurements. XRD and FTIR results showed adsorption of TiO₂ and La₂O₃ nanoparticles on the surface of ZSM-5 support. Photocatalytic activities of the supported catalysts were examined for decolorization and degradation processes of two common reactive dyes: Reactive red 198 (RR198) and Reactive orange 122 (RO122) solutions under catalysts. Incorporation of lanthanum ions improved the photocatalytic activities of TiO₂ supported by ZSM-5 for photocatalytic degradation of the studied reactive dyes under experimental conditions.

INTRODUCTION:

Heterogeneous photocatalysis based on TiO₂ is one of the most successful and convenient method for the quantitative destruction of undesirable contaminants in wastewater.^[1-3] Zeolites are considered to be the useful adsorbent owing to their high surface area, high thermal stability and eco-friendly nature^[4] Moreover, zeolites possess amphoteric properties; Lewis-acidity to denote electron-accepting ability and Lewis basicity to describe electron-donating property. Addition of lanthanum which is an inner transition metal ion (La³⁺) together with TiO₂ and ZSM-5, w improves the zeolite activity because La³⁺ ions have the ability to form complexes with various Lewis bases (e.g. amines, aldehydes, alcohols,

etc.)^[5].

Such complex formation occurs with the interaction of Lewis base and orbitals of La³⁺ ions. La³⁺ ions provide concentrates the organic pollutant at the catalyst surface^[6]. Furthermore, under UV irradiation, the photogenerated electrons transfer from TiO₂ sites to La³⁺ ions, leading to an effective separation of electron-hole pairs^[7].

EXPERIMENTAL:

1-Materials:

The raw ZSM-5 was H⁺ type used as a support in this study was obtained from 2–5 nm crystal size, 0.55 nm pore size, 400 m²/g surface area, Si/Al=1000). The support was characterized by X-ray Diffraction (XRD) analysis.

There is a kind of uniform morphology which has the chemical composition of 97.57% SiO₂ and 1.42% Al₂O₃ was investigated. Titanium tetra-iso-propoxide (98%, Aldrich), acetic acid (96%, Merck), lanthanum nitrate hexa hydrate (99%, Merck), (4-[(4-dimethylamino) phenyl]-azo) benzene sulfonic acid sodium salt (methyl orange, Merck), nitric acid (65%, Merck) and ammonia (NH₃) (25% Merck) were used. Deionized water, purified using Elga-Pure Water Purification system, was used to prepare the used solutions in the experiments.

2-Preparation of catalysts:

TiO₂ was prepared by means of an acid-catalyzed sol-gel method from an alkoxide precursor as follows: 20 ml of titanium tetra isopropoxide was added gradually to 80 wt% acetic acid solution under continuous stirring for 2 h at 50°C to produce a transparent solution. The molar ratio of acetic acid to the alkoxide was 35.2 g of ZSM-5 was suspended in water about 2 h. Depending on the loading of TiO₂ on the support, requisite amount of titania-sol as 5 wt% or 10 wt% was added to the aqueous suspension of ZSM-5. The resulting mixed suspension was agitated by magnetic stirrer for 12 h at room temperature, followed by several centrifugations at (4000 rpm) and washed with deionized water to make the supernatant nearly neutral. The resulting supported catalysts were dried at 100°C for 12 h. The calcination step was done in a muffle furnace using air at 600°C with a heating rate of 10°C min⁻¹ for 5 h. Finally, the catalysts were ground into fine powder, stored in dark and hereafter named as; 2.5-10% TiO₂-ZSM-5.

An ion-exchange process was followed by incorporation of lanthanum ions on the supported catalyst containing the average loading of TiO₂ (e.g., 2.5-10% TiO₂/ZSM-5)^[8]. For this purpose, the contents of lanthanum ions

were regulated as 0.28, 0.5, 1.0 and 3.0 wt% in the final catalyst configuration by solving the requisite amount of lanthanum precursor in de-ionized water. 1.0 g of 7.5% TiO₂-ZSM-5 catalyst was then added to each of these solutions and stirred overnight at room temperature in order to promote the successfully mixing with TiO₂/ZSM-5. Then, the obtained products were centrifuged at 4000 rpm and washed with deionized water several times. The exchange process was repeated four times and the resulting catalysts dried at 100°C for about 12 h and calcined at 600°C with a heating rate of 10°C min⁻¹ for 5 h in a muffle furnace using air. The final product is then ground and stored in the dark and hereafter named as; 0.25% La-7.5% TiO₂-ZSM-5, 0.5% La-7.5% TiO₂-ZSM-5, 1.0% La-7.5% TiO₂-ZSM-5 and 3% La-7.5% TiO₂-ZSM-5.

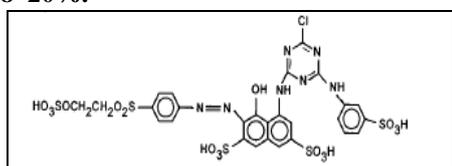
3-Photocatalytic experiments:

The photoactivity experiments were carried out in a cylindrical Pyrex glass reactor containing different loading of catalyst and 250 ml of dye solutions of 100 ppm concentration at 30°C for 60 min. A 30W medium pressure Hg - lamp (254 nm) immersed within the photo reactor was used. The removal efficiency of dye solutions have been calculated by applying the following equation:

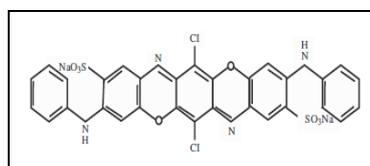
$$\text{Removal efficiency}\% = \frac{[C_0 - C]}{C_0} \times 100$$

Where C₀ is the original dye concentration, C is the retained dye solution. Before all photocatalytic runs, a fresh solution (250 ml) of dye solutions were adjusted to the required pH, and the catalyst was suspended at 0.4 g/l concentrations. Suspensions were kept in dark and magnetically stirred at 30°C for 60 min. The

results indicate that adsorption efficiency was about 18–20%.



(a) Molecular structure of Reactive Red 198 dye



(b) Reactive Orange 122 dye

Fig. (1): Molecular structure of reactive dyes

RESULTS AND DISCUSSION:

1-Characterization of catalysts:

a-X- Ray Diffraction (XRD) analysis:

XRD spectra of 0.25% La-1.0% La-7.5% TiO₂/ZSM-5 catalyst were shown in Fig. (2). In this pattern, formation of La₂O₃ was searched due to the variation of rare earth salts into rare earth oxides during the calcination processes^[9]. Characteristic planes of La₂O₃ are observed at 29.25, 29.94, 45.86, 48.65, 52.03 and 55.438 (2θ), respectively. In such a catalyst system, overlapping of diffraction peaks attributed to TiO₂, La₂O₃ and ZSM-5 support makes the observation of high angle diffractions complicated and not easily detected. Also adsorption of La₂O₃ induces some deformation on the lattice of 7.5% TiO₂/ZSM-5 and accordingly, the crystal volume increases from 5347.78 (Å³) to 5392.39 (Å³) as La% increases from 0.25% to 1.0% (Table 1).

b-Fourier Transform Infra Red (FT-IR) analysis:

Fig. (3) Shows FTIR spectra of raw ZSM-5, 0.25-1.0% TiO₂-ZSM-5 and 0.25-1.0% La-7.5% TiO₂-ZSM-5. In La-7.5% TiO₂-ZSM-5. The O-H stretching modes of surface hydroxyls and adsorbed water molecules are not observed as

absorption peak around 3550 cm⁻¹. This may be due to the successful dehydration of ZSM-5 and supported catalysts at 600°C. The band at 460 cm⁻¹ is due to the bending mode of T-O-T. When TiO₂ is added to ZSM-5 may cause decreasing intensity of the bands at 3550–3630 and 1220–1085 cm⁻¹. Also, the band observed for La (III) atoms on non-framework position was seen at 3685 cm⁻¹. The strongest absorption peak at 1105 cm⁻¹ is assigned to the framework stretching vibration band of Si (Al)-O in tetrahedral Si(Al)O₄ in raw ZSM-5. The position of this peak remains almost unchanged in all supported catalysts, indicating that ZSM-5 structure is not destroyed even after it is loaded with TiO₂ and La₂O₃ nanoparticles. On the other hand, a remarkable decrease in the intensity of the peak at 1105 cm⁻¹ is noticed for 7.5% TiO₂-ZSM-5 catalyst. The increased intensity of this band of La ions measures the extent of perturbation of T-O-T vibrations of the zeolite lattice^[10]. The band at 1060 cm⁻¹ in ZSM-5 shows a shift to 1105 cm⁻¹ indicative for some exchanging Na⁺ by La³⁺ cations. The band at 549 cm⁻¹, assigned to external bonds of the five rings, becomes less intense as La exchanged sodium. Such intensity reduction is normally an indication of crystallinity loss^[11, 12].

Table (1): XRD characterization of La/Ti/ZSM-5 zeolites as a function of increasing La³⁺ loadings where a, b and c are the unit cell (Å) and (V) crystal volume (Å³)

Crystallinity %	V (Å ³)	c	B	a	ZSM-5 type
100%	5334.71	13.375	20.032	19.911	zsm-5 Raw
81.25	5347.78	13.389	20.048	19.923	0.25% La/Ti/zsm-5

67.9	5366.99	13.391	20.118	19.922	0.5% La//Ti/zsm-5
56.8	5372.84	13.405	20.129	19.912	0.75% La/Ti/zsm-5
39.6	5392.39	13.444	20.158	19.898	1.0% La/Ti/zsm-5

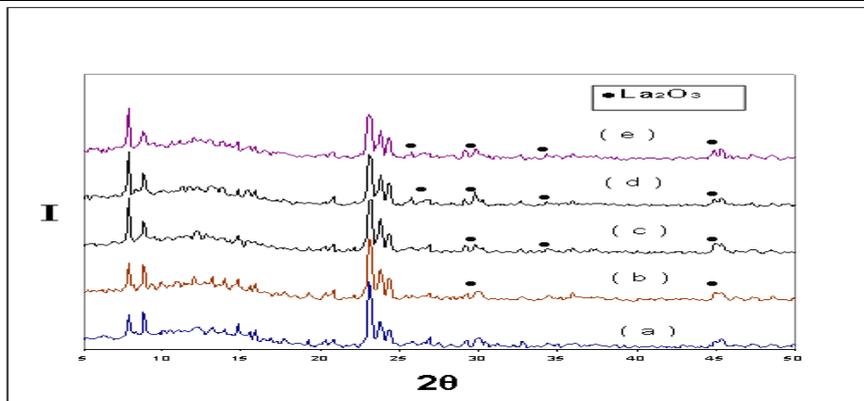


Fig. (2): XRD of : (a)7.5% Ti/ZSM-5, (b) 0.25%La-7.5%Ti/ZSM-5, (c) 0.5%La- 7.5% Ti/ZSM-5, (d) 0.75%La-7.5%Ti/ZSM-and (e) 1.0%La-7.5% Ti/ZSM-5

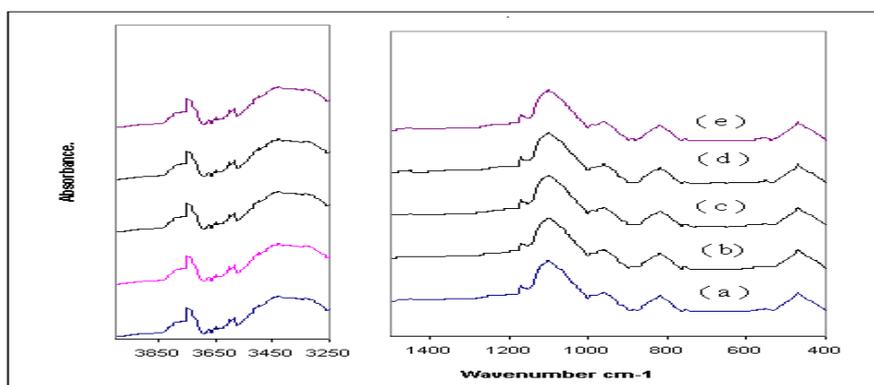


Fig. (3): FT-IR spectra of : (a)7.5%Ti/ZSM-5, (b) 0.25%La-7.5% Ti/ZSM-5, (c) 0.5%La-7.5 %Ti/ZSM-5, (d) 0.75%La-7.5% Ti/ZSM-and (e) 1.0%La-7.5%Ti/ZSM-5

c- UV-Vis DRS analysis:

UV-Vis is a technique to evaluate the presence of the heteroatom in the framework even in such low concentrations. Especially, in the case of transition metal substituted molecular sieves, heteroatom can present either inside the framework or inside the cavity as bulk oxide. The UV-Vis DRS of 7.5% Ti/ZSM-5 and 0.25–1.0% La-7.5%TiO₂/ZSM-5 samples were displayed in Fig. (4). The absorption edges of the supported catalysts are found to shift slightly towards shorter wavelength regions

360–372 nm. The shift decreases with the increase of La³⁺ ion percentage in the final catalyst in comparison to ZSM-5 accordingly, band gap energies of La loaded catalysts are estimated to be 3.3–3.5 eV based on absorption edges of ca. 365–375 nm.

d-Surface texture:

Generally, addition of cations to zeolites by impregnation method increasing the probability of oxide-species formation which blocking the pores of these samples.

It was demonstrated by Lippens and de Boer^[13,14] that for several aluminas and aluminum hydroxides in which the multi-molecular layers of adsorbed nitrogen could be formed freely on all parts of the surface, the amount of adsorbed nitrogen as a function of the surface, the relative pressure is beyond the statistical monolayer, almost independent of the nature of the sample. The thickness of the adsorbed multilayer t (in Å) is found by dividing the volume adsorbed by the surface area according to:

$$t = m \cdot V_{sp} / 22414 \times V_a / S_{BET} \times 10^4 \dots\dots(1)$$

Where V_a is the volume adsorbed (ml STP/g) and S is the surface area (m²/g) calculated from the adsorption isotherm by means of the BET method (Fig. 5). The factor 15.47 is connected with the conversion of the volume of gaseous adsorbate (STP) to liquid volume. Instead of the surface area, S , the volume of gas required to cover the whole surface area of the solid unimolecularly, V_m , can also substitute the factor 3.54 which represents the statistical thickness of a monolayer of adsorbed nitrogen in a multilayer packing. The graph representing the layer thickness as a function of the relative pressure is called t -curve. The experimental values of adsorbed nitrogen which are measured as a function of a relative pressure may be plotted with the aid of the t -curve as a function of the t -values. Hence $V_a = f$

(t) instead of $V_a = f(p/p^0)$. As long as the surface is really accessible; a straight line is obtained which intercepts the origin. Its slope measures the surface area as expressed in equation (1). The t -plot is considered to be the graph of V_{ads} vs. t . If both reference and sample isotherm are identical, as in the case for nonporous solids, a straight line passing through or passing close to the origin, should result. A remarkable decrease of surface area was noticed in the sample 1.0% La/Ti/ZSM-5 ($S_{BET} = 753.5$ m²/g.), (see table 2). Fig. (5) illustrates the downward deviations which were recorded for La/Ti/ZSM-5 samples at $t = 3.2 - 5.5$ Å. Also, Upward deviation were recorded for La/Ti/ZSM-5 at $t = 7.4 - 9.37$ Å due to the existence of wide pores in mesopores structure. As well as determination of the external surface area of micropores solids (S_{ext}^t). Based on the observation that, after the complete filling of the micropores only the adsorption on the external surface influences the experimental isotherm, the slope of the straight line of the plot of $V_a - t$ plot, is directly proportional to the external surface area (S_{ext}^t). The intercept for $t = 0$ gives the volume adsorbate in the micropores (V_{μ}^t)^[15]. Then S_{μ} can be calculated:

$$S_{\mu} = S_{ext} - S_{BET} \dots\dots\dots(2)$$

All surface texture values for La/Ti/ZSM-5 samples are represented in table (2).

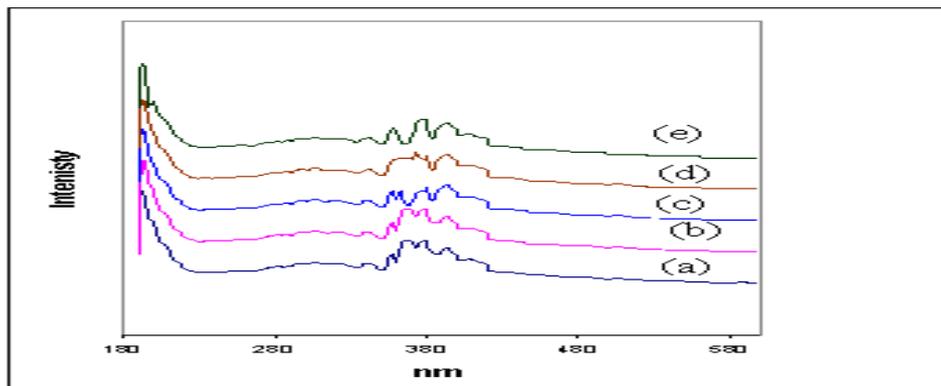


Fig. (4): UV-Vis DRS of (a) 7.5%Ti/ZSM-5, (b) 0.25%La-7.5% Ti/ZSM-5, (c) 0.5%La-7.5 %Ti/ZSM-5, (d) 0.75%La-7.5% Ti/ZSM-5 and (e) 1.0%La-7.5%Ti/ZSM-5

Table (2): Surface texture values for La/Ti/ZSM-5 samples as a microporosity of samples are calculated as sumption of raw ZSM-5 is 100%

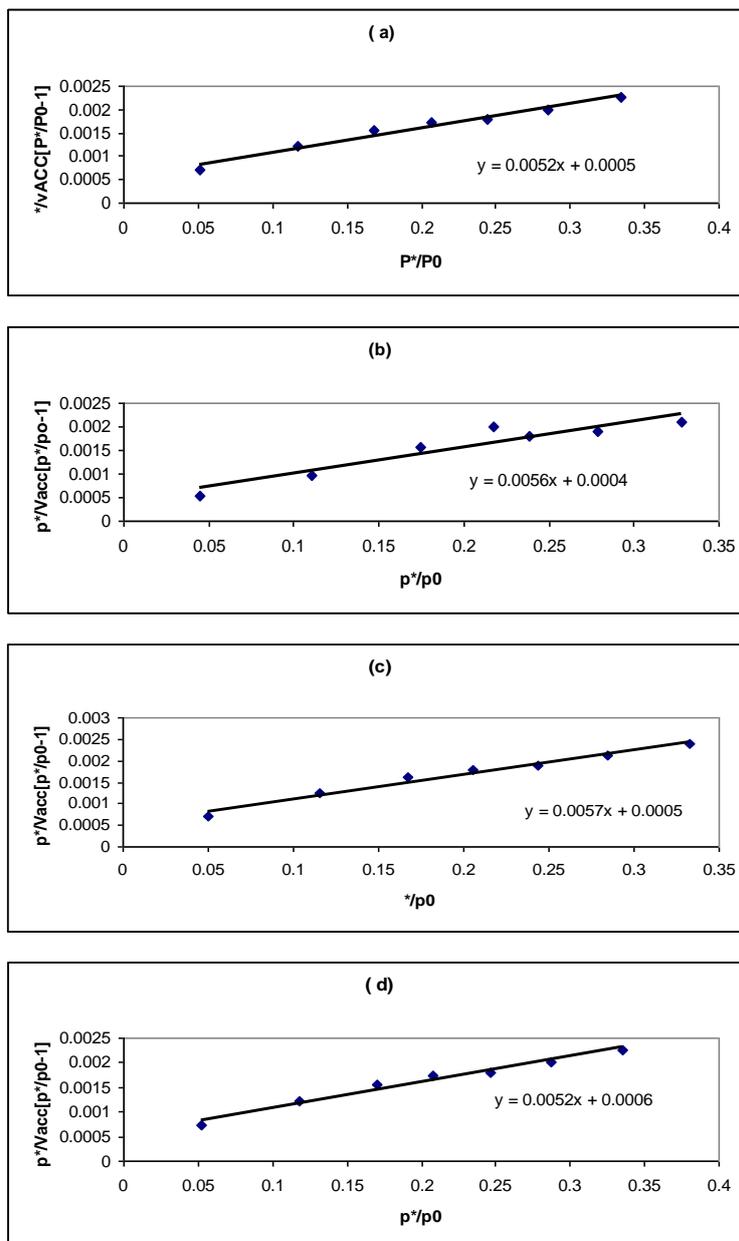


Fig. (5): BET plot of (a) 7.5% Ti/ZSM-5 (b) 0.25% La-7.5% Ti/ZSM-5 (c) 0.5% La-7.5%Ti/ZSM-5 and (d) 0.75%La-7.5%

PHOTOCATALYTIC ACTIVITIES:

1-Adsorption studies:

Preliminary experiments were carried out in dark over 0.25%-1.5% La -7.5% TiO₂-ZSM-5 catalysts. Fifty milligrams of each catalyst in 50 ml of dye (50 ppm) solution is allowed under

stirring in dark. Aliquots were withdrawn at regular intervals and the change in dye concentration is monitored by evaluating the Total Organic Carbon (TOC) which is used as indicator to the presence of organic compounds in solution. The extent of equilibrium adsorption was determined from the decrease in

dye concentration. Maximum adsorption is reached within 60 min for 1.0% La -7.5% Ti/ZSM-5 (19%) but this adsorption decrease at higher La content (16% removal at 1.25% La 7.5% Ti/Z and 18% removal at 1.5% La 7.5% Ti/Z) as shown in Fig. (6). These data can be explained as follows, at lower concentration of lanthanum the probe molecule, RR198 dye is

adsorbed with its SO_3^- and Cl^- groups oriented towards the backbone of the supported catalyst hence existence of La_2O_3 concentrate RR198 molecule on the catalyst surface. At higher concentration of lanthanum, larger ionic radius of La^{3+} ions (103 pm) which hosted within the pores of ZSM-5 structure hinder adsorption of dye molecules.

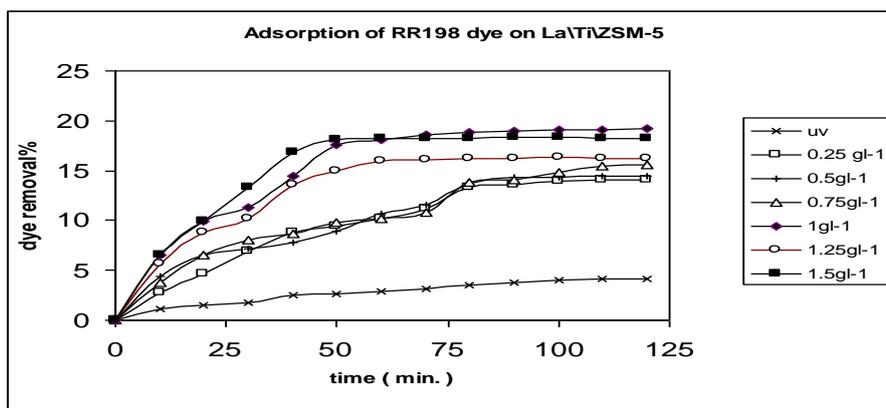


Fig. (6): Adsorption of RR198 dye on different amounts of 0.5% La-7.5% Ti/ZSM-5 as compared to photolysis by UV irradiation

2-Photolysis using UV radiation:

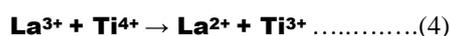
The photolysis experiment is carried out under the UV light by taking 300 ml of dye solution in glass reactor and the results are depicted in Fig. (6). Only 4% of degradation is observed after 10 h of UV irradiation. This indicates a negligible degradation in the presence of UV light without any catalyst. There is no significant degradation over ZSM-5 zeolite and this illustrates that there is no remarkable photocatalytic ability for La/Ti/ZSM-5 and it acts only as a support, Fig. (6).

3-Photocatalytic degradation of dyes using UV/H₂O₂/La/Ti/zsm-5:

a-Effect of lanthanum ion content:

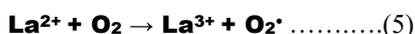
Fig. (7) illustrates the effect of lanthanum ion content on the decolorization and degradation processes of RR198. The lowest

lanthanum ion containing catalyst (0.25% La-7.5% TiO₂/ZSM-5) increases the removal of RR198, whereas further increase in the lanthanum ion loading (more than 1.0% La content) results in decrease in RR198 dye removal percentages. As already mentioned, the lanthanum ion has the ability to increase the concentration of RR198 dye on the surface of the catalyst. Another beneficial of lanthanum ion loading is the inhibition of electron-hole recombination reactions. Science, ZSM-5 acts as a hole trap and La^{3+} ions behave as electron traps according to the following reactions:



However, stability of La^{3+} ions with the electron configuration of [Xe] core is disturbed in the reduced form ($\text{La}^{2+}:[\text{Xe}] 5d1$) with an extra

electron in 5d orbital. For this reason, La^{2+} ions have a tendency to transfer the trapped electron to O_2 according to Eq. (5):



The produced superoxide radical ($\text{O}_2^{\cdot-}$) in Eq. (5) plays a major role in the generation of $\cdot\text{OH}$ radicals by the following reactions:



Oxidative degradation of azo dyes occurs generally by the subsequent attacks of $\cdot\text{OH}$ radicals^[16].

b- Effect of catalyst amount:

In order to determine the optimum amount of catalyst required for effective Photo- catalytic dye degradation, 0.25, 0.5, 0.75, 1.0 and 1.5 g l^{-1} catalyst amounts are evaluated using 1% La 7.5% Ti/ZSM-5 for RR198 dye and the obtained results are shown in Fig. (8). By varying catalyst amounts, it is observed that 0.5 g l^{-1} is found to be optimum for RR198 dye removal. Increasing 0.75–1.5 g l^{-1} , the rate of photocatalytic activity is increased and at higher amounts above these amounts of catalyst (1.0 g l^{-1} -1.5 g l^{-1}) the activity is not found to be encouraging. The successful dye degradation with lower catalyst amount 0.5 g l^{-1} of La/Ti/Z (96.9%) as compared to the corresponding amount (0.5 g l^{-1}) of Ti/ZSM-5 (70.69%) illustrates the effectiveness of lanthanum loaded on dye photodegradation.

c- Effect of H_2O_2 dose:

The effect of H_2O_2 dosage (ranging from 3 cm^3 to 10 cm^3) on the decolorization efficiency was investigated while fixing all the reaction conditions and altering the H_2O_2 doses, Fig. (9).

The conditions of the treatment experiment are listed as follows: UV power 30 Wt, optimum wt of catalyst (1.0 g l^{-1} of 1.0% La-7.5 wt% Ti/ZSM-5 for RR198 dye and 1.5 g l^{-1} of 5 wt% Ti/ZSM-5 for RO122 dye), dye concentration 100 ppm, reaction time 180 min., pH 7.5, volume of the dye solution 300 cm^3 , room temperature. The effect of the H_2O_2 dose on the decolorization efficiency is shown in Fig. (9). It can be observed that the dosage of H_2O_2 required to successful RR 198 dye removal is 8 cm^3 (96.75% at residential time=70 min) as in case of using (UV/ H_2O_2 /Ti/ZSM-5) but with higher degradation efficiency reach to 98.55% at residential time=45 minutes under the same conditions. The high efficiency of RR198 dye removal was observed in comparatively low hydrogen peroxide concentration (8 ml l^{-1}). The fact that the amount of H_2O_2 required for completely photo degradation of reactive dyes by (UV/ H_2O_2 /La/Ti/ZSM-5) can be explained by the Photo catalytic degradation mechanisms^[17].

d-Effect of initial dye concentration:

Incorporation of La^{3+} into Ti/ZSM-5 lead high photodegradation of reactive dyes even at high dye concentrations. The dye concentrations of 50 to 300 ppm are investigated over 1.0 g l^{-1} of 1.0% La- 7.5% Ti/ZSM-5, the results are depicted in Fig. (10). The results show that, by increasing the initial dye concentration from 50 ppm to 300 ppm the removal efficiency decreased from 99.11% to 73.25% in residential time 50 min and 70 min respectively. This can be explained by the fact that, at higher dye concentrations, there would be more adsorption of dye on TiO_2 and La^{3+} resulting in a lesser availability of catalyst surface for hydroxyl radical generations. Joanna and Morawski (2002)^[18] suggested the formation of several layers of adsorbed dye on photocatalyst surface, at higher dye concentrations. Similarly more of

photons would be adsorbed by the dye at higher concentrations which also reduces the energy available for hydroxyl generation. Higher initial concentration also produces more intermediates

which may competitively inhibit the dye degradation. Similar behavior was observed by Neppolian *et al.* (2002)^[19].

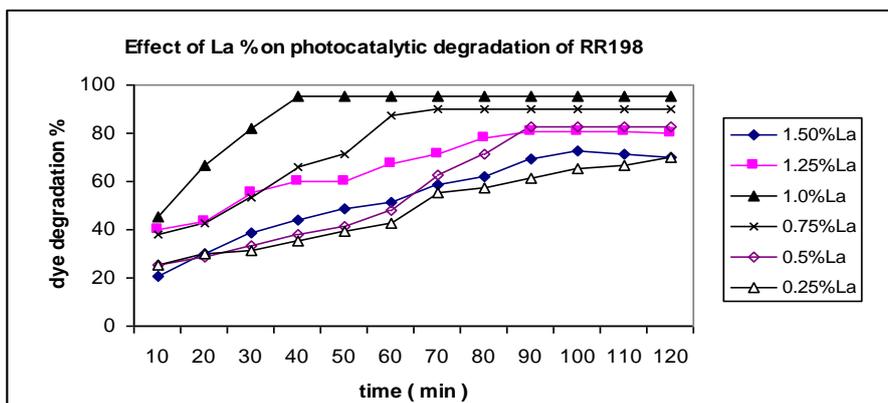


Fig. (7): Effect of La% on RR198 dye degradation by UV/H₂O₂/Ti/ZSM-5

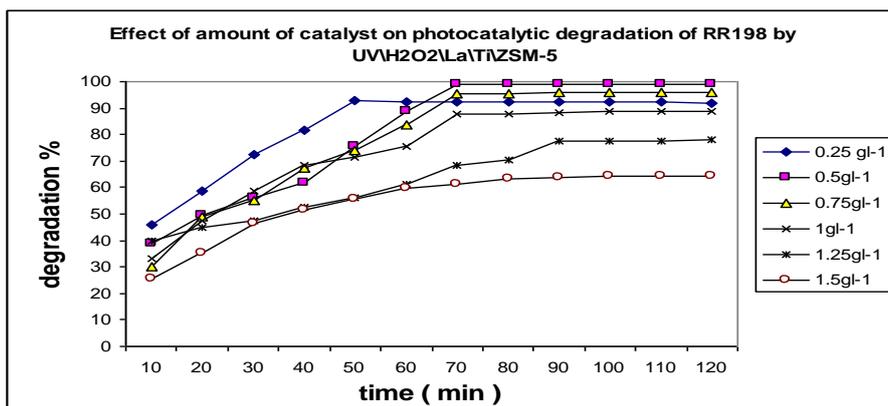


Fig. (8): Effect of amount of catalyst on RR198 dye degradation by UV/H₂O₂/La/Ti/ZSM-5

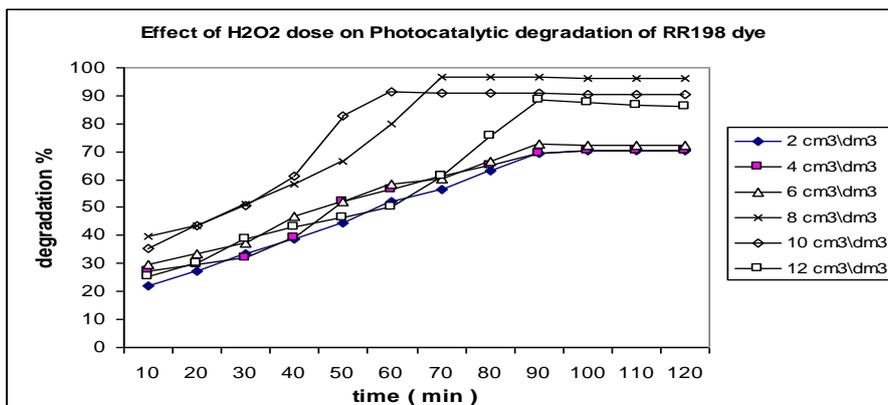
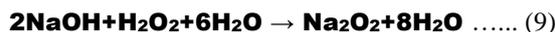


Fig. (9): Effect of H₂O₂ dose on RR198 dye degradation by UV/H₂O₂/La/Ti/ZSM-5

e-Effect of pH:

Fig. (11) illustrates the higher removal efficiency in basic medium (pH 9-11). Since the dye has sulfonic groups in its structure, which is negatively charged, the acidic solution favors adsorption of dye onto catalyst surface, which acquires positive charge in acidic solution. Since the dye has sulfonic groups in its structure, which is negatively charged, the acidic solution favors efficiency of reactive dye removal (99% -100%) in acidic medium (pH=3-6). pH increasing leads to a decrease in the decolorization efficiency. This can be explained by the fact that part of H₂O₂ is used for the

oxidation of alkalis (NaOH) during the decolorization forming sodium peroxide (Na₂O₂). As a result, less hydrogen peroxide is available for the formation of hydroxyl radicals and consequently the degradation of the dye decreases. This behavior is especially noticeable at higher concentration of NaOH as follows:



Also, at higher pH values, the TiO₂ surface is negatively charged and repulsive forces between the TiO₂ surface and dye molecule led to a lower adsorption^[17].

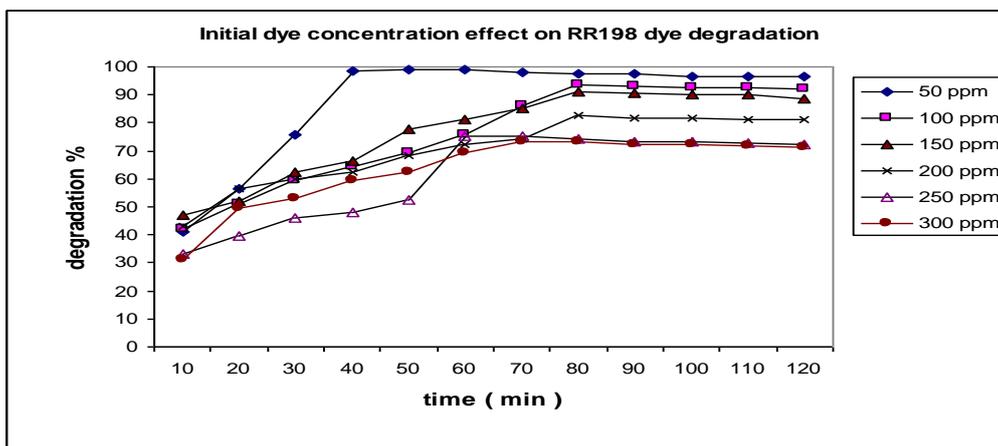


Fig. (10): Effect of initial dye concentration of RR198 dye degradation by UV/H₂O₂/La/Ti/ZSM-5

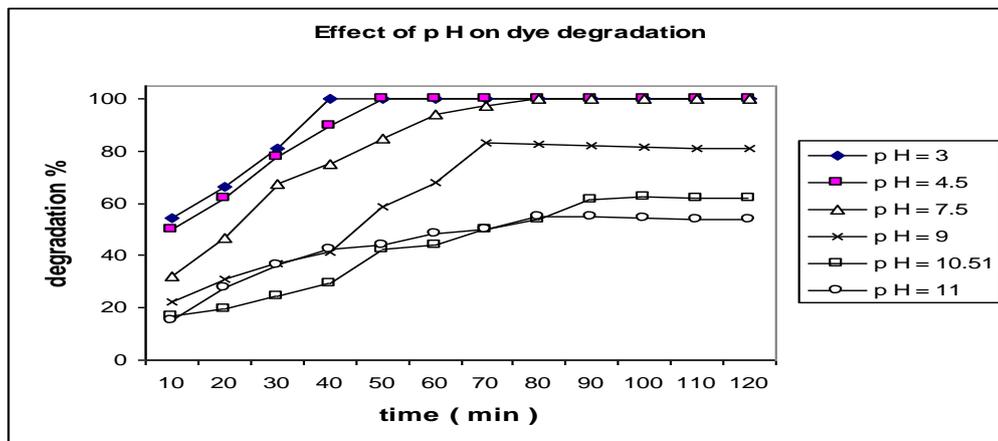


Fig. (11): Effect of pH on RR198 dye degradation using UV/H₂O₂/La/Ti/ZSM-5

CONCLUSION:

Photocatalytic degradation of reactive dyes RR198 and RO122 on La/Ti/ZSM-5 has been achieved successfully. Degradation of reactive dyes is more preferable in acidic medium but can be achieved successfully in neutral medium.

REFERENCES:

- 1-A. Fujishima, T. N. Rao and A. D. Tryk, (2000): *J. Photochem. Photobiol. C: Photochem. Rev.* 1, p. 1.
- 2-M. R. Hofmann, S.T. Martin, and W. Choi, D.W. Bahnemann, (1995): *Chem. Rev.* 95, p. 69.
- 3-M. A. Fox, and M. T. Dulay, (1993): *Chem. Rev.* 93 p. 341.
- 4-H. Yoneyama and T. Torimoto (2000): *Catal. Today* 58 p. 133.
- 5-Y. Zhang, H. Zhang, Y. Xu and Y. Wang, J. Mater (2003): *Chem.* 13 (2003) p. 2261.
- 6-Y. Zhang, H. Xu, Y. Xu, H. Zhang and Y. Wang, (2004): *J. Photochem. Photobiol. A: Chem.* 170, p. 279.
- 7-M. Uzunova-Bujnova, R. Todorovska, D. Dimitrov, and D. Todorovsky, (2008): *J. apsusc*, 05 (2008) p. 331.
- 8-R.T. Yang, N. Tharappiwattananon and R.Q. Long, (1998): *Appl. Catal. B: Environ.* 19, p. 289.
- 9-A. W. Xu, Y. Gao, and H. Q. Liu, (2002): *J. Catal.* 207 p. 151.
- 10- Q. Sun and W.M.H. Sachtler, (2003): *Appl. Catal. B* 42, p. 393
- 11-E. F. Sousa-Aguiar, V. L. Doria-Camorim, F. M. Zanon-Zotin, and Correa dos Santos, (1998): *Microporous Mesoporous Mater.* 25 p. 25.
- 12-E. M. Flanigen, and J. Rabo (Ed.), (1976): *Zeolite Chemistry and Catalysis*, American Chemical Society, Washington, DC, (Ch. 2).
- 13-C.S. Turchi, and D.F. Ollis, (1989): *J. Catal.* 119, 4483.
- 14-S.I. Nishimoto, B. Ohtani, H. Kajiwarra, and T. Kagiya, (1985): *J. Chem. Soc., Faraday Trans.* 181, 61.
- 15-Z. Zhang, C.C. Wang, R. Zakaria, and J. Ying, (1998): *J. Phys. Chem. B* 102, 10871.
- 16-J. T. Spadaro, L. Isabelle, and V. Renganathan, (1994): *Environ. Sci. Technol.* 28 p. 1389.
- 17-A. Neren Ökte and Özge Yılmaz (2009): *Appl. Catal. A: General* 354, p. 132, 99.
- 18-Joanna, G. and Morawski, A.W., (2002): *Applied Catalysis B: Environmental* 36, 45–51.
- 19-B. Neppolian, H. C. Choi, S. Sakthivel, Arabindoo, Banumathi, and V. Murugesan, (2002): *Journal of Hazardous Materials* 89 (2–3), 303–317.

التفكك الحفز ضوئى لبعض الصبغات بواسطة الزيوليت (La/Ti/ZSM-5)

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تهدف هذه الدراسة إلى تفكيك بعض الأصباغ العضوية المستخدمة فى صباغة المنسوجات، وذلك عن طريق تحسين خواص الزيوليت وذلك بإدخال التيتانيوم الرباعى فى صورة الأكسيد بطريقة الادمصاص إلى الزيوليت ثم تطبيق طريقة التفكك الحفز ضوئى. ثم إدخال اللانثانيوم الثلاثى بطريقة التبادل الأيونى ليعطى (La/Ti/ZSM-5). وقد تم التحقق من التركيب النهائى للحافز بعدة تقنيات تتضمن دراسة حيود الأشعة السينية وطيف امتصاص الأشعة تحت الحمراء، وطيف امتصاص الأشعة فوق البنفسجية. كما تم دراسة الخواص السطحية للحافز حيث مساحة السطح وحجم المسام وقطره. وذلك بغرض استخدامه كعامل حفاز فى تكسير بعض أصباغ الازو العضوية النشطة، وهى الصبغ البرتقالى النشط ١٢٢ والصبغ الأحمر النشط ١٩٨ عند تركيزات وظروف تطابق الحالة الموجودة عليها هذين الصبغين فى مياه صرف مصانع الغزل والنسيج. وقد توصلنا إلى النتائج الآتية:-

- ١- التفكك التام لمحلولى الصبغ بنسبه تصل إلى ١٠٠%.
- ٢- الزمن اللازم لإتمام عملية التفكك للأصباغ قصير نسبياً ولا يتجاوز ١٢٠ دقيقة.
- ٣- رغم أن الدراسة أكدت أن الوسط الحمضى مفضل فى عملية التفكك إلا أن عملية تفكك الأصباغ أعطت نسب عالية ومقبولة فى الوسط المتعادل مما يعطى قيمه اقتصادية لهذه الطريقة.