Characterization and photocatalytic activity of SiO$_2$-TiO$_2$ mixed oxide nanoparticles prepared by sol-gel method

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SiO$_2$-TiO$_2$ mixed oxides have been prepared by sol-gel technique from titanium tetraisopropoxide and tetraethylorthosilicate. The prepared materials were characterized by X-ray diffraction, FT-IR spectroscopy, scanning electron microscopy, nitrogen physisorption and UV-vis diffuse reflectance spectroscopy. The results indicate that compared to pure TiO$_2$, the SiO$_2$-TiO$_2$ mixed oxides show higher thermal stability, larger surface area, greater band gap energy and smaller crystallite size. The higher thermal stability of SiO$_2$-TiO$_2$ nanoparticles allows calcination of SiO$_2$-TiO$_2$ nanoparticles at higher temperatures without the accompanying phase transformations from anatase to rutile. The photocatalytic activity of the prepared materials has been compared in the photodegradation of new fuchsin (C.I. 42520) and amaranth (C.I. 16185). Effects of SiO$_2$ concentration and calcination temperature on the photocatalytic activity of SiO$_2$-TiO$_2$ nanoparticles have been investigated. The photodegradation rate of new fuchsin in the presence of 40% SiO$_2$-TiO$_2$ nanoparticles calcined at 600 ºC was higher than that observed with pure TiO$_2$ and Degussa P25 TiO$_2$ by 7.9 and 3.27 times, respectively.

Keywords: Catalysts, Photocatalysts, Mixed oxides, Oxides, Sol-gel method, Photodegradation, Nanocomposites, Titania, Silica

IPC Code: Int. Cl. B01J2/08; B01J21/06

Synthetic dyes are toxic refractory chemicals which can generate intense color and are hazardous to the environment. Most of dyes are resistant to aerobic biodegradation$^1$. Heterogeneous photocatalytic detoxification of water and wastewater contaminants has become one of the promising technologies for waste management, particularly for decomposition of organic pollutants such as dye contaminants$^{2,3}$. Among the metal oxide semiconductors suitable for photocatalytic degradation of environmental pollutants, titanium dioxide (TiO$_2$) is the most widely used catalyst, due to its fascinating physicochemical properties, high photoactivity, photo-corrosion stability, nontoxic and low cost$^{4,6}$. The photocatalytic activity of titanium dioxide can be improved by the addition of SiO$_2$ which increases the available surface area of the catalyst, allowing an increase in adsorption of pollutant molecules. The improved adsorption of pollutant molecules on the surface of silica, in turn improves the photocatalytic activity of SiO$_2$-TiO$_2$ mixed oxides as compared to pure TiO$_2$. Furthermore, the addition of SiO$_2$ increases the amount of surface adsorbed water and hydroxyl groups, which influences the photocatalytic activity of SiO$_2$-TiO$_2$ mixed oxides$^{7,8}$. Properties of SiO$_2$-TiO$_2$ mixed oxides are strongly dependent on the preparation route. The methods used for the preparation of binary oxide nanomaterials include conventional solid state mixing, hydrothermal, co-precipitation and sol-gel processes$^{12,15}$. Amongst these methods, the sol-gel technique is the most effective method for controlling textural and surface characteristics of nanomaterials to produce homogenous materials with a high surface area$^{16-19}$. We report herein the synthesis of SiO$_2$-TiO$_2$ nanoparticles from organometallic titanium tetraisopropoxide (TTIP) and tetraethylorthosilicate (TEOS) by sol-gel method. The prepared materials have been characterized by X-ray diffraction, scanning electron microscopy, energy dispersive spectroscopy, FT-IR, nitrogen physisorption and UV-vis diffuse reflectance spectroscopy. The extensive use of amaranth (C.I. 16185) and new fuchsin (C.I. 42520) in textile and cosmetics industries and also difficulty in their biodegradation make them appropriate candidates for photodegradation experiments. Therefore, the photocatalytic activity of the resultant materials has been compared in the photodegradation of new fuchsin (cationic dye) and amaranth (anionic dye). Also, the effects of SiO$_2$ concentration and calcination temperature on the photocatalytic activity of prepared materials have been evaluated.
Materials and Methods
Preparation and characterization of photocatalyst nanoparticles

All chemicals used in the synthesis of photocatalysts were analytical grade (Merck, Germany) and used without further purification. New fuchsin (I) and amaranth (II) were purchased from Acros Organics and used as model pollutants in photocatalytic activity measurements.

SiO$_2$-TiO$_2$ mixed oxides were prepared by the sol-gel method as follows:

TTIP and TEOS were used as precursors for TiO$_2$ and SiO$_2$, respectively. Ethanol and isopropanol (IPA) were used as mutual solvents, and nitric acid was used as a catalyst for hydrolysis. The molar ratios of H$_2$O, EtOH, HNO$_3$ and IPA to total alkoxides (TEOS + TTIP) were 150:1 : 0.2:4, respectively. An acidic aqueous solution was prepared by the addition of HNO$_3$ (65 wt. %) to H$_2$O. To the resultant solution, TEOS was added dropwise under vigorous stirring. In a separate flask, TTIP was added to a mixture of IPA and EtOH. The obtained alcoholic solution was added to the aqueous solution of TEOS under vigorous stirring and was refluxed for 6 h at 85 ºC. Then, a white precipitate was obtained. The precipitate was filtered, washed sequentially with ethanol and doubly distilled water, dried at 60ºC and calcined at 400, 500, 600, 800 and 950ºC for 3 h.

XRD patterns of prepared samples were recorded with a Siemens D5000 diffractometer using Cu-K$_\alpha$ ($\lambda = 0.154056$ nm) radiation. The average crystallite size of prepared particles was determined by the Scherrer\textsuperscript{20} equation, $D$=0.9$\lambda$/$\beta$cos$\theta$, where $D$ is the average crystallite size (nm), $\lambda$ is the wavelength of X-ray radiation, $\beta$ is the full-width at half-maximum of the diffraction peak and $\theta$ is the diffraction angle.

Morphology and chemical composition studies were carried out using a Philips XL-30 scanning electron microscope equipped with an EDS apparatus. The structure of prepared materials was investigated with an FT-IR spectrometer (Bruker Tensor 27) using KBr as a background. Optical band gap energies of prepared materials were determined with UV-vis diffuse reflectance spectroscopy using an Avantes AvaSpec-2048 TEC fiber optics spectrometer. Nitrogen adsorption-desorption studies were performed at 77 K on a Belsorp mini II apparatus. Specific surface area and pore size distribution were determined using BET and BJH, respectively.

Photocatalytic activity
Photocatalytic activity was studied at room temperature using a 100-mL quartz tubular batch photoreactor placed vertically in front of a UV lamp (15 W, $\lambda_{max} = 254$ nm, manufactured by Philips, Netherlands). In each experiment, 0.1 g of powdered photocatalyst was uniformly dispersed in 100 mL of an aqueous solution of new fuchsin (10 mg L$^{-1}$) or amaranth (20 mg L$^{-1}$) by sonication for 15 min (Elma Transonic T460/H). A uniform suspension was transferred into the reactor, and a stream of oxygen was passed through the reactor at a flow rate of 0.6 mL min$^{-1}$. Prior to irradiation, the reaction suspension was equilibrated for 30 min in darkness to achieve adsorption-desorption equilibrium of the reactant solution and catalyst particles. Next, the UV lamp was turned on and the photocatalytic reaction was allowed to begin. Throughout the photocatalytic reaction, 5 mL of solution was taken at defined intervals and centrifuged to remove the catalyst prior to analysis. The residual concentration of new fuchsin or amaranth was monitored using a UV-vis spectrophotometer (Pharmacia Biotech Ultrospec 2000, England) at 553 or 522 nm, according to the maximum absorbance wavelengths of new fuchsin and amaranth, respectively. The activity of the

<table>
<thead>
<tr>
<th>C.I. No.</th>
<th>$\lambda_{max}$ (nm)</th>
<th>Mol. wt. (g mol$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>42520</td>
<td>553</td>
<td>365.91</td>
</tr>
<tr>
<td>16185</td>
<td>522</td>
<td>604.46</td>
</tr>
</tbody>
</table>

New fuchsin (I)  
Amaranth (II)
prepared photocatalysts in the photodegradation of new fuchsin and amaranth was compared to the activity of commercial Degussa P25 TiO₂.

Results and Discussion

Characterization of nanocatalysts

XRD patterns of SiO₂-TiO₂ mixed oxides with different concentrations of SiO₂ calcined at 500 ºC are shown in Fig. 1(a), while XRD patterns of 40 % SiO₂-TiO₂ mixed oxides calcined at different temperatures are illustrated in Fig. 1(b). XRD analysis results indicate that, even when the calcination temperature was increased to 950 ºC, the major crystalline phase of the prepared particles was exclusively anatase, without any rutile, and the SiO₂ was formed as an amorphous phase. This result confirms that the addition of SiO₂ to TiO₂ increases the thermal stability of TiO₂ crystallites, resulting in the suppression of phase transformation from anatase to rutile. The suppression of anatase-rutile transformation was due to the presence of SiO₂, which prevented the nucleation of rutile by impeding the direct contact (mutual contact) of titania particles through the formation of the Ti—O—Si linkage²¹. However, further addition of SiO₂ into TiO₂ matrix decreases the crystallinity of anatase TiO₂ and thus, 50 % SiO₂-TiO₂ mixed oxides display an amorphous structure. This phenomenon is attributed to the formation of Si—O networks by excessive SiO₂, which prevents the production of anatase crystallites²². The average crystallite size of the prepared particles is given in Table 1. XRD patterns show broader diffraction peaks for SiO₂-TiO₂ mixed oxides than for the pure TiO₂, indicating that SiO₂-TiO₂ mixed oxides crystallites are smaller than the pure TiO₂ crystallites. The crystallites of pure TiO₂ calcined at 500ºC were 11.4 nm, while those of 40 % SiO₂-TiO₂ mixed oxides calcined at the same temperature were 6.7 nm. Thus, the addition of amorphous SiO₂ to TiO₂ hinders the growth of anatase TiO₂ crystallites. Additionally, calcination temperature had a strong effect on the size of crystalline TiO₂ particles. The crystallite size of anatase TiO₂ increased from 5.7 to 15.1 nm with an increase in calcination temperature from 400 to 950 ºC.

SEM micrographs and EDS spectra of pure TiO₂ (Fig. 2 (a)) and 40 % SiO₂-TiO₂ nanoparticles calcined at 500 ºC (Fig. 2 (b)) show the formation of aggregated secondary particles by the agglomeration of primary particles. Additionally, the SiO₂-TiO₂

Fig. 1—XRD patterns of (a) pure TiO₂ and SiO₂-TiO₂ nanoparticles with different concentrations of SiO₂ calcined at 500 ºC, and, (b) 40% SiO₂-TiO₂ nanoparticles calcined at different temperatures.
agglomerates possess a rough and porous surface, resulting in an increased surface area. The chemical composition of the samples was analyzed by EDS. The results indicated that the chemical composition of 40% SiO$_2$-TiO$_2$ was 40.43 at. % Si and 59.57 at. % Ti, confirming that TiO$_2$ is well distributed in a SiO$_2$ monolith. This composition concurs with theoretical calculations.

FT-IR spectra of pure TiO$_2$ and 40% SiO$_2$-TiO$_2$ calcined at 500 ºC shows bands at 802 and 1084 cm$^{-1}$, assigned to symmetric and asymmetric Si–O–Si stretching vibrations, respectively. The band corresponding to asymmetric Ti–O–Si stretching vibrations is located at 962 cm$^{-1}$, indicating that Ti is present in four-fold coordination with oxygen from SiO$_4$ structure$^{23}$. The peak at 1626 cm$^{-1}$ is attributed to bending vibrations of O–H and is ascribed to chemisorbed water, while the broad peak at 3415 cm$^{-1}$ may be attributed to O–H stretching vibrations. The XRD and FT-IR results reveal that SiO$_2$ exists as a segregated amorphous phase in an anatase TiO$_2$ matrix, with portions of Ti–O–Si bonds$^{24}$.

Figure 3 shows UV-vis DRS spectra of the prepared materials with different amounts of SiO$_2$ and UV-vis DRS spectra of 40 % SiO$_2$-TiO$_2$ mixed oxides calcined at different temperatures. The reflectance data were converted to the Kubelka-Munk function$^{25}$, $F(R)$, according to the following equation, $F(R)=(1-R)^2/2R=k/s$, where $F(R)$ is the Kubelka-Munk function, $R$ is the reflectance, and $k$ and $s$ are Kubelka-Munk absorption and scattering coefficients, respectively. As shown in Fig. 3, band gap energies can be derived from UV-vis spectra by plotting $(F(R)h\nu)^2$ versus photon energy (h$\nu$). Calculated band gap energies reported in Table 1 shows that the addition of SiO$_2$ to TiO$_2$ increased the optical band gap energy of SiO$_2$-TiO$_2$ mixed oxides. This phenomenon is

Table 1—Crystalline phase, average crystallite size and optical band gap energies of the prepared nanoparticles

<table>
<thead>
<tr>
<th>Sample</th>
<th>Calcination temp. (ºC)</th>
<th>Crystallite phase</th>
<th>Crystallite size (nm)</th>
<th>Band gap (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure TiO$_2$</td>
<td>500</td>
<td>100% Anatase</td>
<td>11.4</td>
<td>3.2</td>
</tr>
<tr>
<td>10% SiO$_2$-TiO$_2$</td>
<td>500</td>
<td>100% Anatase</td>
<td>8</td>
<td>NA</td>
</tr>
<tr>
<td>20% SiO$_2$-TiO$_2$</td>
<td>500</td>
<td>100% Anatase</td>
<td>7.7</td>
<td>3.33</td>
</tr>
<tr>
<td>30% SiO$_2$-TiO$_2$</td>
<td>500</td>
<td>100% Anatase</td>
<td>7</td>
<td>NA</td>
</tr>
<tr>
<td>40% SiO$_2$-TiO$_2$</td>
<td>500</td>
<td>100% Anatase</td>
<td>6.7</td>
<td>3.35</td>
</tr>
<tr>
<td>50% SiO$_2$-TiO$_2$</td>
<td>500</td>
<td>Amorphous</td>
<td>N/A</td>
<td>NA</td>
</tr>
<tr>
<td>40% SiO$_2$-TiO$_2$</td>
<td>400</td>
<td>100% Anatase</td>
<td>5.7</td>
<td>NA</td>
</tr>
<tr>
<td>60% SiO$_2$-TiO$_2$</td>
<td>800</td>
<td>100% Anatase</td>
<td>11.7</td>
<td>3.33</td>
</tr>
<tr>
<td>80% SiO$_2$-TiO$_2$</td>
<td>950</td>
<td>100% Anatase</td>
<td>15.1</td>
<td>3.3</td>
</tr>
</tbody>
</table>

NA = Not analyzed
N/A = Not applicable

Fig. 2—SEM micrographs and EDS spectra of (a) pure TiO$_2$, and, (b) 40% SiO$_2$-TiO$_2$ nanoparticles.

Fig. 3—Kubelka-Munk transformed reflectance spectra for (a) pure TiO$_2$ and SiO$_2$-TiO$_2$ nanoparticles with different concentrations of SiO$_2$ [1, pure TiO$_2$; 2, 20% SiO$_2$; 3, 40% SiO$_2$], and, (b) 40% SiO$_2$-TiO$_2$ nanoparticles calcined at different temperatures [1, 500 ºC; 2, 600 ºC; 3, 800 ºC; 4, 950 ºC].
attributed to the quantum size effect\textsuperscript{26}, which is caused by a decrease in the crystallite size of anatase TiO\textsubscript{2}, from 11.4 nm in pure TiO\textsubscript{2} to 6.7 nm in 40\% SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides. In contrast, the optical band gap energy of SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides gradually decreased as the calcination temperature increased. Thus, by increasing the calcination temperature from 500 to 950 °C, the band gap energy decreased from 3.35 to 3.30 eV, and the crystallite size increased from 6.7 to 15.1 nm. Due to the greater band gap energy of SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides as compared to that of pure TiO\textsubscript{2}, the SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides are expected to possess a higher activity for photooxidation and photo-reduction than pure TiO\textsubscript{2}.

Nitrogen adsorption-desorption isotherms of pure TiO\textsubscript{2} and 40 \% SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides at 77 K are shown in Fig. 4(a). According to IUPAC classification, pure TiO\textsubscript{2} displays a type IV isotherm and H2 hysteresis, indicating a mesoporous material that exhibits capillary condensation and evaporation\textsuperscript{27}. Similarly, 40\% SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides were characterized by type III adsorption isotherms, indicating mesoporous and macroporous materials\textsuperscript{28}. The specific surface area of 40 \% SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxide (106.13 m\textsuperscript{2} g\textsuperscript{-1}) was significantly higher than that of pure TiO\textsubscript{2} (46.87 m\textsuperscript{2} g\textsuperscript{-1}). Pore size distribution was determined by the BJH model and shows that pure TiO\textsubscript{2} possessed a narrow bimodal distribution (1-6 nm) (Fig. 4 (b)). However, pore size distribution in SiO\textsubscript{2}-TiO\textsubscript{2} was broader (1-60 nm), which is consistent with the mesoporous and mesoporous-macroporous structures of pure TiO\textsubscript{2} and SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides, respectively. According to the BJH analysis, the addition of 40 \% SiO\textsubscript{2} to TiO\textsubscript{2} increased the pore volume from 0.063 cm\textsuperscript{3} g\textsuperscript{-1} in pure TiO\textsubscript{2} to 0.256 cm\textsuperscript{3} g\textsuperscript{-1} in 40\% SiO\textsubscript{2}-TiO\textsubscript{2} mixed oxides. The high porosity and larger pore size facilitate the mass transfer of reactants such as oxygen and reaction intermediates, which improves photocatalytic activity\textsuperscript{24}.

Photocatalytic activity studies

The photoactivity of the prepared materials in the photodegradation of two organic dyes, viz., new fuchsin and amaranth, was investigated. As mentioned above, in all experiments, prior to irradiation the suspension of the photocatalyst in the dye solution was stirred in dark for 30 min to establish adsorption-desorption equilibrium. The concentration of the dye at this point was used as the initial concentration (C\textsubscript{0}) for further kinetics studies. The kinetics of the photocatalytic degradation process has been well described by the Langmuir-Hinshelwood\textsuperscript{5} kinetic model (Eq. 1),

\[
\frac{dC}{dt} = \frac{kKC}{1 + KC} \quad \text{... (1)}
\]

where \( r \) is the photodegradation rate (mg L\textsuperscript{-1} min\textsuperscript{-1}), \( k \) is the reaction rate constant (mg L\textsuperscript{-1} min\textsuperscript{-1}), \( K \) is the equilibrium adsorption coefficient of the reactant (L mg\textsuperscript{-1}), \( t \) is the irradiation time (min) and \( C \) is the concentration of the reactant at time \( t \) (mg L\textsuperscript{-1}).

When the initial concentration of the reactant (C\textsubscript{0}) is low, integration of Eq. 1 yields the simplified pseudo-first-order kinetics equation (Eq. 2),

\[
\ln \left( \frac{C_0}{C} \right) = k_{app}t \quad \text{... (2)}
\]

where \( k_{app} \) is the apparent pseudo-first-order rate constant (min\textsuperscript{-1}) and \( C_0 \) is the initial concentration of the reactant (mg L\textsuperscript{-1}) (after establishment of adsorption-desorption equilibrium). The pseudo-first-
order rate constant has been chosen as the basic kinetic parameter to compare different catalysts photocatalytic activity. Since it is not related to the adsorbed dye concentration, it is possible to determine the photocatalytic activity independent of the previous adsorption period in the dark.

The photolytic experiments with new fuchsin and amaranth were carried out in the absence of the photocatalysts. These experiments indicate that only a small amount of dyes were photolyzed, and therefore, the direct photolysis of dyes without photocatalysts can be considered to be negligible, with less than 2 % conversion within 1 h of the UV irradiation.

The effect of SiO₂ concentration and calcination temperature on the photodegradation of new fuchsin was investigated. Photocatalytic activity of the SiO₂-TiO₂ mixed oxides as a function of SiO₂ concentration is shown in Fig. 5(a) and the values of apparent rate constants ($k_{app}$) are given in Table 2. These results indicate that the photocatalytic activity of SiO₂-TiO₂ mixed oxides increases significantly with an increase in SiO₂ content up to 40 mol %. According to XRD, BET, BJH, FT-IR and DRS results, the superior photocatalytic activity of SiO₂-TiO₂ mixed oxides is due to the smaller particle size, higher surface area, larger pore volume, more surface hydroxyl groups and greater band gap energy of SiO₂-TiO₂ mixed oxides. However, a SiO₂ concentration greater than 40 mol % decreased the photocatalytic activity of SiO₂-TiO₂ mixed oxides due to a decrease in TiO₂ concentration and crystallinity. Photodegradation of new fuchsin as a function of calcination temperature in the presence of 40% SiO₂-TiO₂ mixed oxides is illustrated in Fig. 5(b). The photocatalytic activity of SiO₂-TiO₂ mixed oxides increased significantly with an increase in calcination temperature and reached a maximum at a calcination temperature of 600 ºC. The increase in photocatalytic activity with increasing calcination temperature is attributed to a higher amount of crystalline anatase TiO₂ and a reduction in the amount of bulk defects, which act as electron-hole recombination centers. However, at calcination temperatures greater than 600 ºC, photocatalytic activity significantly decreased due to an increase in particle size and decrease in band gap energy. The maximum photocatalytic activity, attained with 40 % SiO₂-TiO₂ mixed oxides calcined at 600 ºC, is 7.9 and 3.27 times greater than pure, synthetic TiO₂ and Degussa P25 TiO₂, respectively.

The effect of SiO₂ concentration on photocatalytic activity of the SiO₂-TiO₂ mixed oxides (calcined at 500 ºC) in the photodegradation of amaranth, an anionic dye, was compared to that of pure TiO₂ and Degussa P25 TiO₂ (Fig. 6). The results indicate that the photocatalytic activity of SiO₂-TiO₂ mixed oxides is significantly lower than that of pure TiO₂ and Degussa P25 TiO₂ (Table 2). Several studies have

![Fig. 5—Effect of (a) SiO₂ concentration [1, P25 TiO₂; 2, pure TiO₂; 3, 10% SiO₂; 4, 20% SiO₂; 5, 30% SiO₂; 6, 40% SiO₂; 7, 50% SiO₂], and (b) calcination temperature [1, 400 ºC; 2, 500 ºC; 3, 600 ºC; 4, 800 ºC; 5, 950 ºC] on the photocatalytic activity of SiO₂-TiO₂ mixed oxides in the photodegradation of new fuchsin.](image-url)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Calcination temp. (ºC)</th>
<th>$k_{app}$ (min⁻¹)</th>
<th>New fuchsin</th>
<th>Amaranth</th>
</tr>
</thead>
<tbody>
<tr>
<td>P25 TiO₂</td>
<td>NC</td>
<td>0.1041</td>
<td>0.4536</td>
<td></td>
</tr>
<tr>
<td>Pure TiO₂</td>
<td>500</td>
<td>0.0432</td>
<td>0.1014</td>
<td></td>
</tr>
<tr>
<td>10% SiO₂-TiO₂</td>
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<td>0.2081</td>
<td>0.0384</td>
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</tr>
<tr>
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<td>0.0327</td>
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<tr>
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<td>0.2646</td>
<td>0.0255</td>
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<td>500</td>
<td>0.2877</td>
<td>0.02</td>
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<tr>
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<td>500</td>
<td>0.0512</td>
<td>0.0037</td>
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</tr>
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<td>400</td>
<td>0.138</td>
<td>NA</td>
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<tr>
<td>40% SiO₂-TiO₂</td>
<td>600</td>
<td>0.341</td>
<td>NA</td>
<td></td>
</tr>
<tr>
<td>40% SiO₂-TiO₂</td>
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<td>0.2454</td>
<td>NA</td>
<td></td>
</tr>
<tr>
<td>40% SiO₂-TiO₂</td>
<td>950</td>
<td>0.0704</td>
<td>NA</td>
<td></td>
</tr>
</tbody>
</table>

NC = Not calcined
NA = Not analyzed
reported that the addition of SiO₂ into TiO₂ reduces the isoelectric point of SiO₂-TiO₂ mixed oxides from a pH of 6.2 for pure TiO₂ to a pH of 2-3 for SiO₂-TiO₂ mixed oxides. A reduction in the isoelectric point of SiO₂-TiO₂ mixed oxides has been attributed to the presence of acidic groups on the catalyst surface. Thus, the surface of the catalyst is negatively charged in a solution with a pH > 3 and positively charged at a pH < 3. The pH of a solution of 20 mg L⁻¹ amaranth is 4.9. Thus, lower adsorption of amaranth on the surface of SiO₂-TiO₂ mixed oxides resulted from electrostatic repulsion between the negatively charged surface of SiO₂-TiO₂ and the organic anion. Photocatalytic degradation of dyes is associated with dye adsorption onto the surface of SiO₂-TiO₂. Furthermore, photodegradation occurs at or near the surface of the catalyst rather than in the bulk solution. The lower photocatalytic activity of SiO₂-TiO₂ mixed oxides in the photodegradation of amaranth is consistent with lower adsorption of amaranth on the surface of the SiO₂-TiO₂ photocatalyst. Hence, it can be concluded that the differences in the photodegradation constants of new fuchsin and amaranth are in accordance with their different adsorptions on the surface of the photocatalyst.

**Conclusions**

SiO₂-TiO₂ mixed oxides have been prepared by the sol-gel method using TTIP and TEOS. XRD results indicate that the addition of SiO₂ into TiO₂ inhibits phase transformation from anatase to rutile and prevents the growth of TiO₂ crystallites. The greater thermal stability of SiO₂-TiO₂ mixed oxides allowed a higher calcination temperature, resulting in anatase TiO₂ with a higher degree of crystallinity and fewer bulk defects, without formation of a rutile phase. FT-IR spectra indicate the presence of Ti–O–Si cross-links and revealed interactions between TiO₂ and SiO₂ at a molecular scale. Ti–O–Si bonds and interactions may enhance surface properties and photoactivity. According to the BET and DRS results, embedding amorphous SiO₂ into a TiO₂ matrix increases the surface area, pore volume and optical band gap energy by suppressing TiO₂ crystallite growth. Increased photocatalytic activity of SiO₂-TiO₂ mixed oxides in the photodegradation of new fuchsin has been obtained by increasing SiO₂ contents up to 40 mol %. The photodegradation rate of new fuchsin with a 40% SiO₂-TiO₂ catalyst calcined at 600 °C is higher than that of pure TiO₂ and Degussa P25 TiO₂ by 7.9 and 3.27 times, respectively.

**References**
