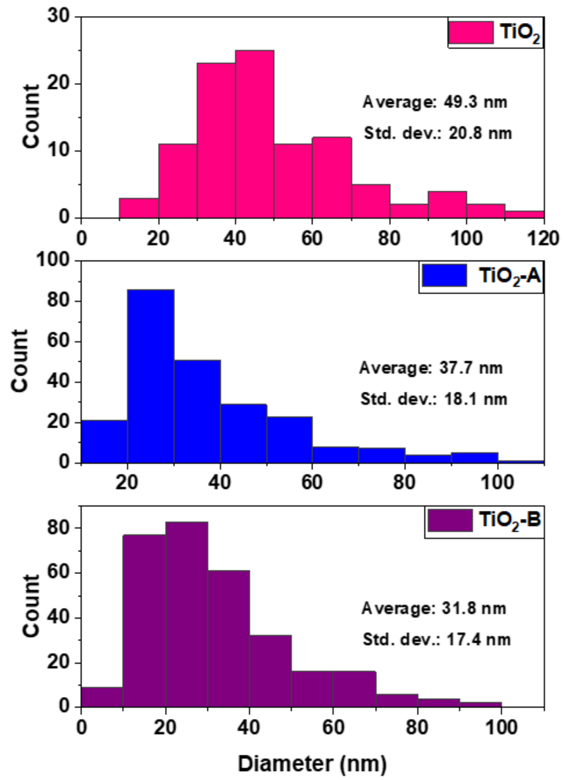
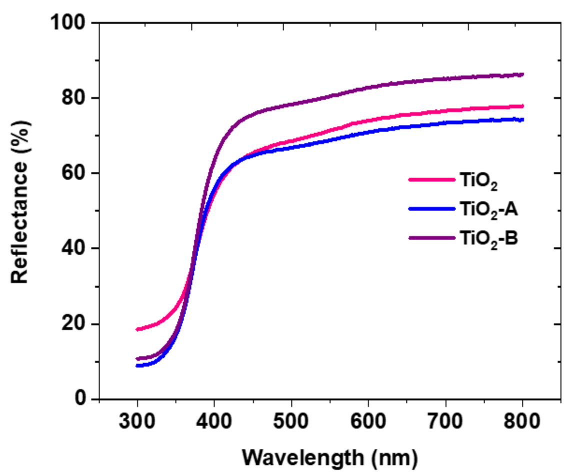
respectively [31]. The $n$ value is 2 for pure and doped $TiO_2$ nanoparticles, ascribed to a permissible indirect transition mode [32]. According to Figure 6, the band gap energies were 2.94 eV, 3.08 eV, and 3.10 eV for $TiO_2$ nanoparticles $TiO_2$-A and $TiO_2$-B, respectively.

**Figure 4. Particle size distribution histogram of the nanomaterials studied.**



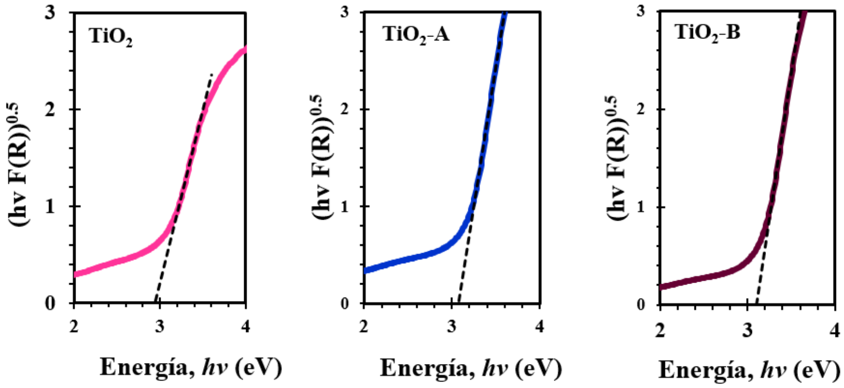

**Source: Authors' own creation.**

**Figure 5. Diffuse reflectance spectrum for TiO2, TiO2-A (0.5 Ag at%), and TiO2-B (0.75 Ag at%).**




**Source: Authors' own creation.**

**Figure 6. Optical band gap energy estimation (Wood and Tauc method) for TiO2, TiO2-A (0.5 Ag at%), and TiO2-B (0.75 Ag at%) nanoparticles.**




**Source: Authors' own creation.**

Currently, the literature reports low bandgap energies of $TiO_2$ nanoparticles doped with Ag ions obtained from different synthesis methods as a result of an electron-hole recombination delay due to the energy states located above the valence band [33]. In this study, the bandgap energy values were significantly higher (see *Table 1*), attributed to the reduction of the electron-hole recombination delay. However, there was no difference between the value for $TiO_2$ nanoparticles compared to $TiO_2$-A and $TiO_2$-B, since the photodeposition method reduced the optical properties through the reduction of $Ag^+$ ions to metallic Ag [19].

**Table 1. Results were reported in recent research related to $TiO_2$ doping with Ag to improve optical properties.**

| Synthesis method | Doping amount | Bandgap reduction (eV) | Reference |
|---|---|---|---|
| Green synthesis and photodeposition/reduction | 0.5 at% and 0.75 at% | Negligible | This study |
| Biomediated doping (BMD) | 1.0 wt.% | 0.70 eV | [34] |
| Sol-gel method | 2 mol% to 8 mol% | 0.67 eV | [35] |
| | 1 wt.%, 3 wt.% and 5 wt.% | 0.32 eV | [36] |
| | 1.5 wt.% | 0.3 eV | [37] |
| | 0.10 mol.% | 0.08 eV | [38] |
| Controlled and energy efficient microwave assisted method | 0.12 mol% to 0.5 mol% | 0.22 eV | [18] |
| Sol-gel/solvothermal (SGH) | 0.04 Ag/Ti molar ratio | 0.25 eV | [39] |
| Photodeposition method | 1.0 wt.% | 0.29 eV | [22] |

**Source: Authors' own creation.**

## Photocatalytic degradation

The direct photolysis process was performed to determine the sensitivity degree of a 40 mg/L acetaminophen solution exposed to solar irradiation in the absence of the catalyst until a minimum equivalent radiation of 8,000 $J/m^2$ was reached. *Equation 6* was used to calculate the degradation percentage in the photolysis, adsorption, and heterogeneous photocatalysis processes [40].

$$Degradation\ (\%) = \frac{C_o - C_t}{C_o} \times 100 \quad (6)$$

where $C_o$ is the initial concentration (mg/L) and $C_t$ is the concentration as a function of time/cumulative radiation. In the results, the photolysis process achieved a degradation rate of 14.5%, showing that the pollutant is slightly susceptible to self-degrade due to exposure to solar radiation. This is an advantage since it represents a savings in terms of photocatalyst material and increases the overall rate during the photocatalytic process. This self-degradation percentage was similar to that reported in other research, which found a reduction of 12% of acetaminophen after irradiation with a mercury lamp (500 W) for 60 min with an initial concentration of 50 mg/L [41].

On the other hand, *Table 2* shows the results obtained from the adsorption experiments avoiding exposure to natural light. Adsorption of acetaminophen with the commercial photocatalyst P-25 was found to be negligible, whereas $TiO_2$ nanoparticles synthesized using the *C. citratus* extract and $TiO_2$-B (0.75 Ag at%) nanostructures showed higher removal percentages due to improvements in their surface area properties. A total surface area of 64.4 $m^2/g$ for $TiO_2$ nanoparticles synthesized via green chemistry using *C. citratus* extract has been previously reported [32], which is 41% higher than that reported for commercial P-25 (45.7 $m^2/g$) [42]. Moreover, an increased photocatalyst dose decreases the adsorption removal percentage attributed to the surface interaction between the nanoparticles (aggregation), leading to a reduction in the number of available adsorption sites [43], [44].

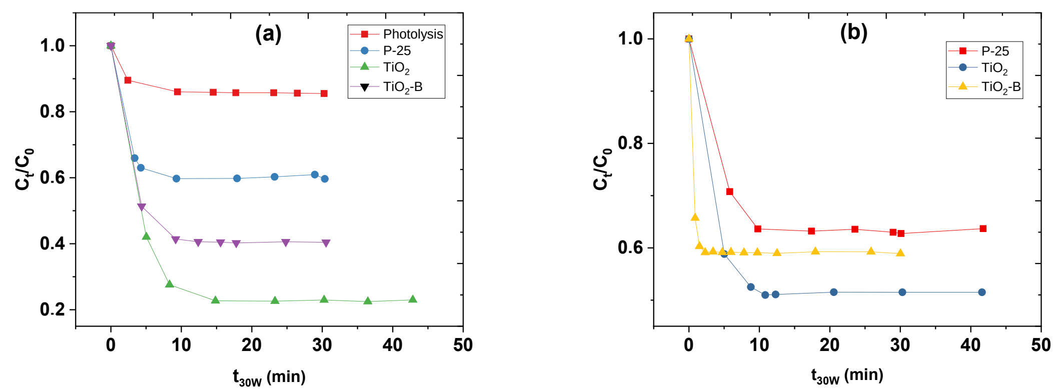
**Table 2. Molecular adsorption percentage of acetaminophen avoiding exposure to natural light and at pH 3 and 40 mg/L after 180 min (equilibrium time).**

| Material | Catalyst dosage | |
|---|---|---|
| | 0.2 g/L | 0.3 g/L |
| Commercial P-25 | 1.7% | 1.4% |
| Green $TiO_2$ nanoparticles | 12.7% | 7.4% |
| $TiO_2$-B (0.75 Ag at%) | 14.3% | 6.7% |

**Source: Authors' own creation.**

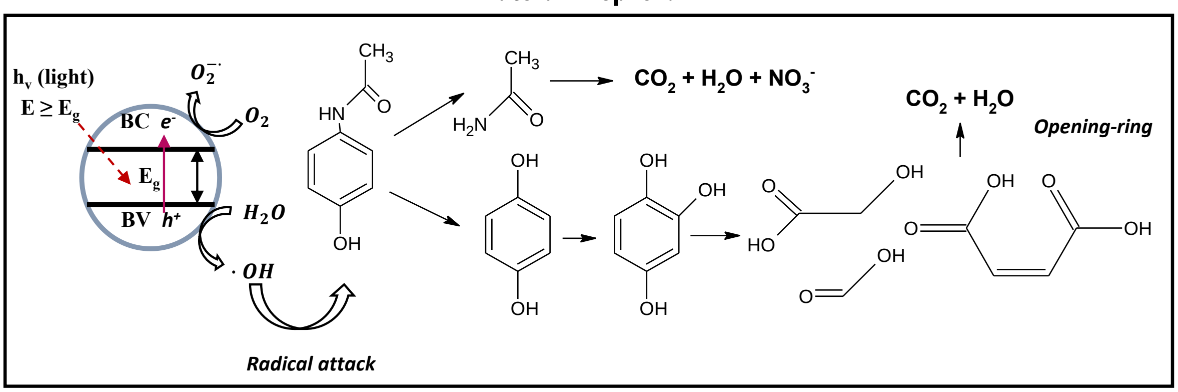
The photodegradation tests were performed on consecutive days and at the same time to reduce the variation due to the environmental conditions. *Figure 7a,b* shows the experimental results of acetaminophen photodegradation using photocatalyst doses of 0.2 g/L and 0.3 g/L of commercial P-25, $TiO_2$ nanoparticles, and $TiO_2$-B nanostructures. Additionally, the behavior of the relative $C_t/C_0$ concentration was evaluated regarding the standardized radiation time ($t_{30W}$). At both photocatalyst doses, commercial P-25 presented the lowest degradation efficiency attributed to the adsorption capacity of acetaminophen, whereas $TiO_2$-B (0.75 Ag at%) showed an intermediate performance due to the photodeposition of metallic Ag ions. In this case, the reduction of $Ag^+$ ions to metallic Ag promotes surface poisoning of the photocatalyst, leading to a reduction in the photocatalytic activity compared to $TiO_2$ nanoparticles synthesized via green chemistry. Moreover, the reaction rate decreases considering the high concentration of the photocatalyst (0.3 g/L), which was attributed to a shielding effect from the reduced light irradiation in the innermost part of the reactor [45], [46].

**Figure 7. Remaining acetaminophen as a function of the standardized radiation time ($t_{30W}$) using (a) 0.2 g/L and (b) 0.3 g/L photocatalyst doses.**




**Source: Authors' own creation.**

**Figure 8. Proposed mechanism by adapting the reported pathways of the photocatalytic degradation of acetaminophen.**




**Source: Authors' own creation.**

The mechanism for the photodegradation of acetaminophen is displayed in *Figure 8*, which was based on information previously reported in the literature [47, 48]. The mechanism initially consisted of the electron/hollow pair ($e^-/h^+$) generation from the activation of the photocatalyst using simulated or natural UV–Vis radiation, as detailed in *Equations 7-12*. Then, the hydroxyl radicals attack the ortho- or para-positions of the aromatic ring in the acetaminophen structure to produce N-Methyl formamide ($C_2H_5NO$), hydroquinone, and 1,4-benzoquinone. Finally, the opening ring/ring cleavage and the oxidation of species, such as maleic acid ($C_4H_4O_4$), formic acid ($CH_2O_2$), and glycolic acid ($C_2H_4O_3$), are generated until their mineralization ($CO_2$ and $H_2O$).

$$TiO_2 + hv \rightarrow h^+ + e^- \tag{7}$$

$$h^+ + OH^-(surface) \rightarrow OH\cdot \tag{8}$$

$$h^+ + OH^-(adsorbed) \rightarrow OH\cdot + H^+ \tag{9}$$

$$e^- + O_2\,(adsorbed) \rightarrow O_2^{-\cdot} \tag{10}$$

$$O_2^{-\cdot} + H^+ \rightarrow HO_2^{\cdot} \tag{11}$$

$$HO_2^{\cdot} + HO_2^- \rightarrow OH\cdot + OH^- + O_2 \tag{12}$$

## Conclusions

This study reports the green synthesis of titanium dioxide ($TiO_2$) nanoparticles using a *Cymbopogon citratus* (*C. citratus*) extract as a capping agent and surface modification with metallic Ag via the photodeposition method. Although *C. citratus* acted as a capping agent to control the growth and particle size, the $TiO_2$ nanoparticles exhibited polycrystallinity attributed to a strong surface effect, leading to the formation of agglomerates with large diameter sizes. The large agglomerates reduce the available active sites for the adsorption of naphthalene since its removal is expected from the photocatalytic degradation instead. On the other hand, the reduction from Ag ions to metallic Ag on the surface of the $TiO_2$ nanoparticles promoted a direct effect on the optical properties, showing no differences compared to the unmodified $TiO_2$ nanoparticles. Although this effect caused surface poisoning on the photocatalyst, decreasing the photodegradation of acetaminophen, the results showed a better performance compared to commercial P-25. Therefore, the green synthesis of $TiO_2$ nanoparticles and photodeposition of metallic Ag on the surface provides an enhanced photocatalytic activity toward the degradation of organic pollutants, in which further research work can involve optimization aiming at better optical properties.

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