Journal of Materials and Environmental Science
ISSN : 2028-2508
e-ISSN : 2737-890X
CODEN : JMESCN

J. Mater. Environ. Sci., 2024, Volume 15, Issue 8, Page 1174-1184

http://www.jmaterenvironsci.com

Photocatalytic degradation of dye from TiO_2 supported on clay beads: case of safranin

SORO Donafologo Baba [1*], N'GUETTIA Kossonou Roland [1], YAYA Coulibaly [2], MEITE Ladji [1], ABOUA Kouassi Narcisse [1], TRAORE Karim Sory**

[1]Laboratory of Environmental Sciences, UFR of Environmental Sciences and Management, NANGUI ABROGOUA University, 02 BP 801 Abidjan 02, Ivory Coast [2]Department of Chemical Engineering, Jilali University, P. M. B. 5025, [2]UFR-Agriculture, Fisheries Resources and Agro-industry, University of SAN-PEDRO, 01 BP 1800 SAN-PEDRO 01, Ivory Coast

** Corresponding author, Email address : baba_soro@yahoo.fr*
*** Corresponding author, Email address : rolandkossonou@yahoo.fr*

Received *30 July 2024,*
Revised *26 Aug 2024,*
Accepted *30 Aug 2024*

Keywords:
- ✓ *Photocatalysis,*
- ✓ *clay, Safranin,*
- ✓ *Adsorption,*
- ✓ *Titanium dioxide;*

Citation*: Soro D.B., N'guettia K.R., Yaya C., Meite L., Aboua K.N., Traore K.S. (2024) Photocatalytic degradation of dye from TiO_2 supported on clay beads: case of safranin, J. Mater. Environ. Sci., 15(8), 1174-1184*

Abstract: This work aims to test the photocatalytic efficiency of TiO_2 fixed on a clay support in the elimination of safranin in an aqueous medium. The clay was taken from the Haut Sassandra region in Ivory Coast. Three types of clay balls were prepared: pure clay balls (simple clay), clay balls impregnated with titanium dioxide (Clay+TiO_2) and balls containing a mixture of clay and TiO_2 (Clay/TiO_2). The beads were calcined at 500 °C in a Nabertherm muffle furnace for 2 hours to make them resistant to water. The efficiency of adsorption and photocatalysis processes was tested for the removal of safranin. The photocatalytic tests were carried out with a polychromatic lamp. The residual concentrations of the dye were measured by UV-visible spectrophotometry. The adsorption results showed after 420 min (7 h) of contact, elimination rates of approximately 43 %, 40 % and 57 % respectively with the simple clay, the clay impregnated with TiO_2 and the clay/TiO_2 for a concentration of 7 mg/L and a clay mass of 5 g. Under the same conditions, more satisfactory results were obtained with the materials (clay+TiO_2) and (clay/TiO_2) during the photocatalysis experiments with respective dye removal rates of 68 % and 76 %. This study suggests that the photocatalytic degradation process using clay combined with TiO_2 could be promising for the remediation of colored water.

1. Introduction

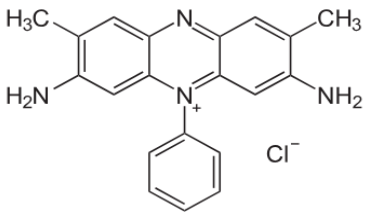
Environmental pollution caused by industrial waste discharges, such as synthetic dyes, represents a serious global problem (Albuquerque *et al.*, 2021) . These synthetic dyes are toxic and their presence in the hydrosphere can have dangerous consequences for aquatic life, reducing the penetration of sunlight and inhibiting certain photochemical reactions (Yusuf *et al.*, 2017) . Several wastewater treatment technologies have been developed, including physical, chemical and biological methods, to reduce the concentration of dyes in industrial wastewater before discharge into water bodies (Krępska *et al.*, 2016; Jodeh *et al.*, 2018; Krishna *et al.*, 2021; Aaddouz *et al.*, 2023; Abu Rub *et al.*, 2023). The textile, leather tanning, cosmetics, pigment and many other sectors industries rely heavily on dyes as the main raw material (Mohan *et al.*, 2008; Tkaczyk *et al.*, 2020), but poor management waste leads to water pollution (Krishna *et al.*, 2021). Among the most used dyes, safranin is a water-soluble

cationic dye with wide applications in the textile and pharmaceutical industries, but can severely damage the environment and human health (Elsherif *et al.*, 2021; Naghipour *et al.*, 2023; Ahmed *et al.*, 2023). Common treatment methods for these effluents include aerobic or anaerobic bioremediation, chemical oxidation, ion exchange, ozonation, Fenton reagent, membrane filtration, electrokinetic coagulation, irradiation and electrochemical degradation (Garg *et al.*, 2019). Although researchers have developed various dye removal methods, the limited performance of these techniques requires the development of new low-cost dye removal approaches (Peramune *et al.*, 2022). The search for innovative treatment processes, both economical and environmentally friendly, therefore constitutes a major challenge in the field of water treatment. Photocatalysis using suspended titanium dioxide (TiO_2) has shown promise for the degradation of chemical contaminants present in aqueous media (Gaya and Abdullah, 2008; Chong *et al.*, 2010). However, its application is limited by the presence of TiO_2 particles in suspension at the end of the treatment. Thus, the use of natural materials such as clay for the adsorption of dyes has attracted increasing interest in the field of contaminated water treatment (García *et al.*, 2023). The coupling of adsorption and photocatalysis seems to be a promising approach to optimize these two techniques in the treatment of dyes. The development of new photocatalytic materials aimed at improving the performance of existing catalysts in the near ultraviolet and visible light represents a major challenge for fundamental research. The objective of this study is to evaluate the photocatalytic efficiency of TiO_2 fixed on a clay support in the elimination of safranin in aqueous media.

2. Methodology

2.1 Chemical products

The commercial "safranin" solution of purity (99 %) used is marketed by Sigma-Aldrich. Concentrated sulfuric acid (H_2SO_4) with a purity of 95-98 %, as well as acetone with a purity of 100 %, come from Prolabo. The dilution solvent used is 99% purity ethanol. All solutions were prepared with distilled water obtained from a distiller. Titanium dioxide (TiO_2) comes from Degussa.



2.2 Origin of clay material

The clay used comes from the Haut-Sassandra region, located in the center-west of Côte d'Ivoire, more precisely from the town of Daloa. It was taken from the bottom of a well and then transported to the laboratory.

2.3 Preparation of clay materials

2.3.1. Simple clay balls

The process of manufacturing clay balls is described in two stages (**figure 1**). The raw clay was first ground in a porcelain mortar to obtain a powder. This was then sieved on a Retsch brand electric sieve. The fraction of clay with a diameter between 45 µm and 125 µm was selected to manufacture the beads (Step 1). Beads of approximately 0.3 cm in diameter were shaped by hand, then dried in an oven at 105 °C for two days. They then underwent calcination at 500 °C for 2 hours to eliminate all residues of organic matter (Step 2).

2.3.2. TiO_2 coated beads

A portion of the previously calcined beads (Step 2) was impregnated in a solution of 0.5 g/L of TiO_2 for 24 hours so that their entire surface was completely covered. They were then calcined at 400 °C for 2 hours (Step 3) (**Figure 2**).

Figure 1. Simple clay ball manufacturing process

Figure 2. Manufacturing process for TiO_2 coated beads

2.3.2. Beads containing TiO_2

The beads containing TiO_2 were made using a clay-TiO_2 ratio of 50 %. For this, 50 g of clay powder were mixed with 50 g of TiO_2 powder (Step 1). Then, 40 mL of distilled water was added to this mixture to form a mixed clay/TiO_2 paste. Balls of approximately 0.3 cm in diameter were shaped from this paste then dried in an oven at 105°C for two days. They were then calcined in the oven at 400 °C for 2 hours (Step 2) (**Figure 3**).

Figure 3. Manufacturing process for beads containing TiO_2

2.4. Experiences

2.4.1. Adsorption tests

2.4.1.1. Adsorption rate

The results expressed in terms of adsorption rate (% adsorption) of safranin on the clay balls are obtained by Eqn . 1 :

$$\text{Adsorption rate (\%)} = \frac{C_i - C_f}{C_i} \times 100 \qquad \text{Eqn . 1}$$

with: C_i : initial concentration of safranin (mg/L) , C_f : residual concentration at equilibrium (mg/L)

2.4.1. 2. Quantity adsorbed

The quantity of safranin adsorbed expressed in mg/g of solute per gram of adsorbent solid is given by Eqn . 2 :

$$Q_t = \frac{(C_0 - C_1)}{m} * V \qquad \text{Eqn . 2}$$

Q_t : Quantity of safranin adsorbed per unit mass of clay (in mg/g), C_0 : Initial concentration of safranin (mg/L), C_1 : Residual concentration at equilibrium (mg/L), V: Volume of the adsorbate (L), m: mass of clay (g)

2.4.1.3. Effect of contact time

5 g of each type of clay beads are placed in mini-reactors containing 50 mL of the safranin solution with a concentration of 7 mg/L. Samples are taken at regular time intervals. The samples taken are analyzed with a UV-visible spectrophotometer to measure the residual concentration.

2.4.1.4. Effect of clay ball mass

Adsorption tests were carried out in mini-reactors containing 50 mL of safranin solution with a concentration of 7 mg/L to which different masses of clay beads (2 to 10 g) were added for a contact time of 420 min (7 hrs). Samples are taken and analyzed with a spectrophotometer.

2.4.1.5. Effect of initial concentration

Different safranin solutions of varying concentrations (1, 3, 5, 7 and 8 mg/L) were prepared in 50 mL mini-reactors at room temperature and at the pH of the solution. To each solution is added a mass of clay balls optimal for a contact time of 420 min (7 h). Samples are taken and analyzed with a spectrophotometer.

2.4.2. Photocatalysis tests

The irradiation device used is a ramp equipped with a light source under which 50 mL quartz mini-reactors are placed. The polychromatic lamp with a wavelength $\lambda > 285$ nm is positioned 20 cm above the mini-reactors. Samples are taken at precise time intervals (0; 30; 60; 120; 180; 240; 300, 360 and 420 min) and analyzed with a UV-visible spectrophotometer to measure the residual concentration.

2.4. 3. UV-visible spectrophotometry

The discoloration of the safranin solutions was monitored using an AQUALYTIC 800 brand UV-visible spectrophotometer (AL 800). The maximum absorption wavelength of safranin was determined to be 520 nm.

3. Results and Discussion

3.1 Adsorption kinetics of safranin on clay beads

3.1.1. Effect of contact time

Figure 4 shows the adsorption of safranin on the three types of materials. These results indicate that after a contact time of 360 min, an equilibrium phase is obtained corresponding to the saturation of the sites available on the surface of the materials. At this equilibrium time, a greater adsorption is observed with the clay/TiO_2 material (53 %) followed by the simple clay (43 %) and the material (clay + TiO_2) (40 %). The increase in the adsorption rate with clay beads containing TiO_2 could be explained by the presence of TiO_2 in the structure of the clay which increases the number of active sites (Manova *et al*., 2010 ; Asfaram *et al*., 2017) . The low elimination rate obtained with the clay balls soaked in the TiO_2 solution would be due to the occupation by the TiO_2 particles of the attachment sites on the surface of the balls, thus reducing the number of sites available for the safranin molecule.

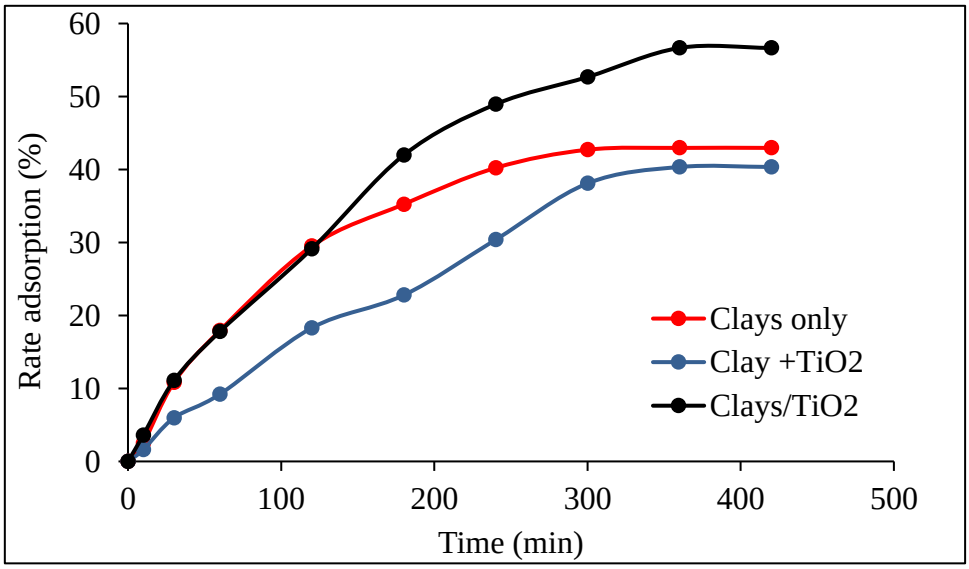


Figure 4. Adsorption kinetics of safranin on different materials
mass = 5 g, Co = 7 mg/L, T = 25 °C

Figure 5 shows the quantities of the dye adsorbed on the different materials. The quantities adsorbed on the simple clay balls and those soaked in the TiO_2 solution are approximately 0.03 mg/g slightly below those obtained with the clay/TiO_2 material (0.04 mg/g). These results are in accordance with those obtained previously. There is therefore a limit beyond which increasing the contact time no longer leads to a significant increase in adsorption capacity (Fayazi *et al*., 2015). Once all adsorption sites of the material are occupied by the dye molecules, there is no further improvement in adsorption kinetics. Therefore, it is essential to determine the optimal adsorption time in order to achieve maximum efficiency while minimizing processing costs (Ain *et al*., 2020 ; Liu *et al*., 2023).

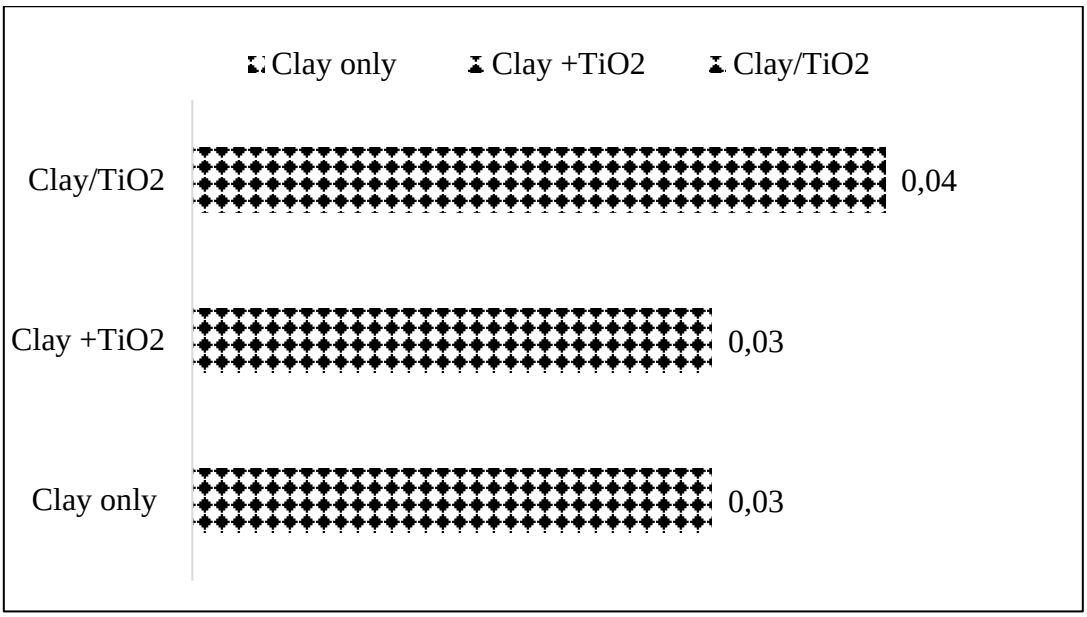


Figure 5. Quantity of safranin adsorbed on the different materials
m = 5 g, Co = 7 mg/L, T = 25 °C

3.1.2. Effect of clay ball mass

The evolution of the safranin adsorption rate as a function of variation in the mass of the clay beads is illustrated by **Figure 6**. An increase in the dye removal rate is observed with increasing mass of each material. Thus, for a mass of 10 g, adsorption rates of 71 %, 72 % and 60 % were obtained respectively on simple clay, clay/TiO_2 and clay+TiO_2 beads. These results could be explained by the fact that the increase in the mass of the adsorbent leads to an increase in the adsorption sites and the surface area brought into contact (Harrou *et al*., 2020 ; Mundkur *et al*., 2022).

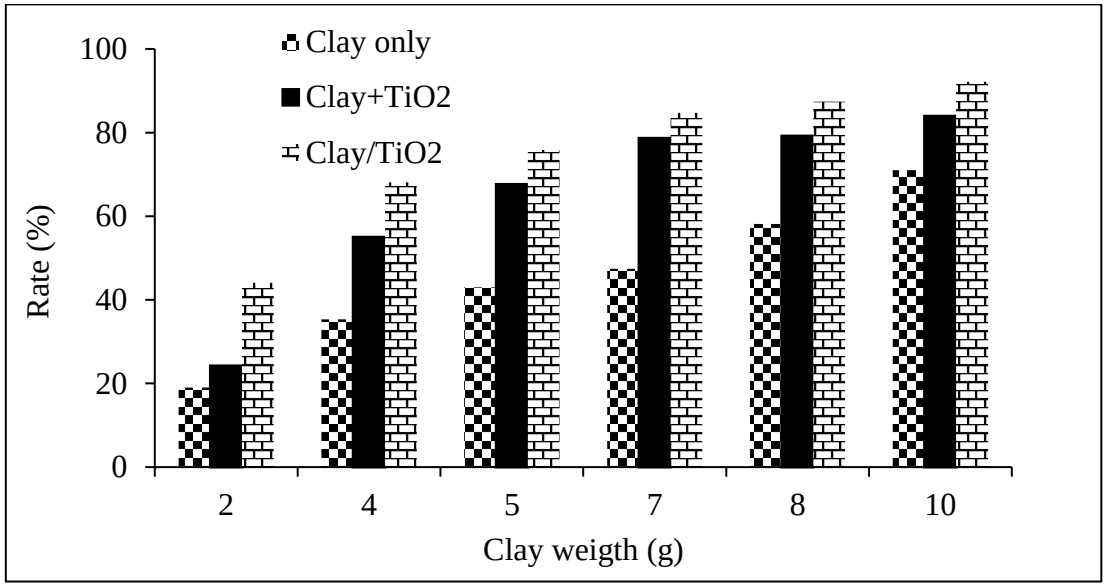


Figure 6 . Adoption rate of safranin depending on mass materials ; Co = 7 mg/L, T = 25°C

3.1.3. Effect of initial concentration

Figure 7 shows the evolution of the percentage of dye removal as a function of concentration. It can be seen that the adsorption rate increases with the initial concentration up to a dye concentration of 3 mg/L for all three materials. These results could be explained by the fact that the increase in the initial

concentration provides a significant driving force which accelerates the diffusion of the molecule across the surface of the adsorbent (Dhafir *et al.*, 2014 ; Awad *et al.*, 2019). Beyond this concentration, the adsorption rate decreases, thus showing saturation of the sites available on the surface of the materials (Bulut and Aydın, 2006 ; Paul *et al.*, 2012). The dye removal percentages obtained at this concentration (3 mg/L) were approximately 47.40 %, 42.46 % and 57 % respectively for simple clay beads, clay beads + TiO_2 and clay/TiO_2 balls.

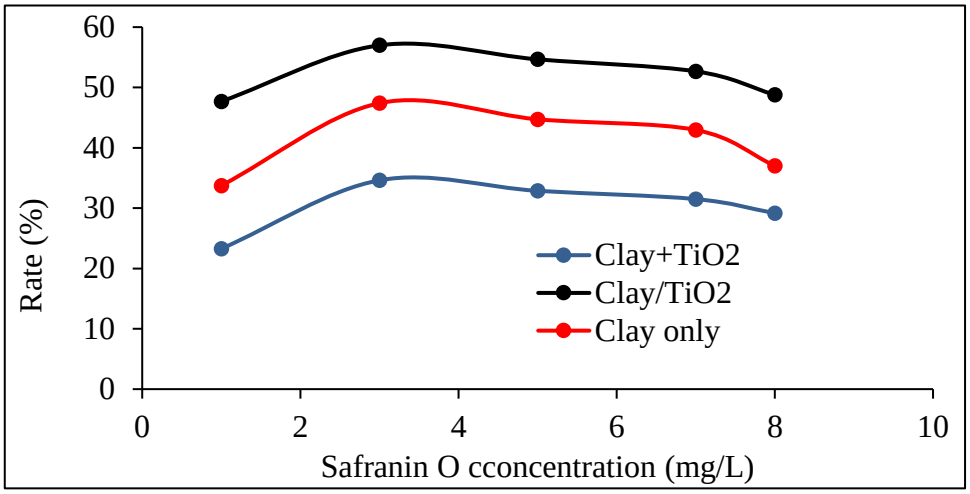


Figure 7. Safranin adsorption rate as a function of initial concentration m = 5 g, T = 25 °C

3.2. Photocatalysis tests

3.2.1. Kinetics of photocatalytic degradation

Two safranin solutions with a concentration of 7 mg/L, one of which containing the material (Clay+TiO_2) and the other the material (Clay/TiO_2), were irradiated under a polychromatic lamp of wavelength $\lambda > 285$ nm. The results are presented in **Figure 8**. This figure indicates that after 420 min (7 h) of irradiation, approximately 68 and 76 % of the dye were degraded by the (Clay+TiO_2) and (Clay/TiO_2) processes, respectively.

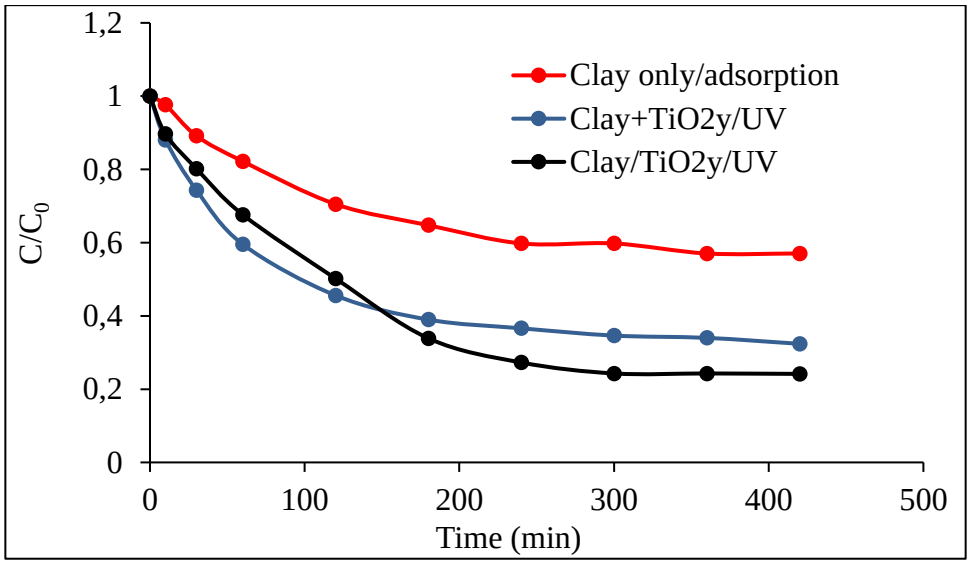


Figure 8. Kinetics of safranin degradation in the presence of composites clay+TiO_2 and Clay/TiO_2 ; C_0 = 7 mg/L, m = 5 g, T = 25 °C

The degradation speed is therefore greater in the presence of the material (Clay/TiO_2). But in both cases, we observe a greater elimination of safranin compared to adsorption on simple clay (43 %). These results reveal that these hybrid materials improve photocatalytic activity. In fact, the dispersion of TiO_2 on the clay surface increases active sites around adsorbed molecules, which increases the interaction with the photo-produced active species making degradation faster (Bencherai and Boumendjel, 2013**).** Based on previous work, one of the common effects that has been observed during photocatalyst integration in clays is the bandgap increment of the composite compared to the intact photocatalyst (Peng *et al.*, 2016 ; Sohrabnezhad *et al.*, 2016). These authors used the $LaFeO_3$ /montmorillonite composite for the degradation of Rhodamine B (RhB) under visible light irradiation. For this composite, the bandgap energy was 2.24 eV, higher than that of FeO_3 alone (2.15 eV).

3.2.2. Influence of parameters on the kinetics of safranin degradation

3.2.2.1. Effect of mass of composite material

Figure 9 illustrates the evolution of the degradation kinetics of the dye as a function of the mass of the composite material. The degradation rate increases with the mass of the hybrid material. Degradation rates of approximately 84 and 92 % were obtained respectively with the clay+TiO_2 and clay/TiO_2 material for a mass of 10 g. The discoloration is greater and faster in the presence of the clay/TiO_2 mixture. These results show that mixing clay with titanium dioxide makes the surface of the material more photo-reactive.

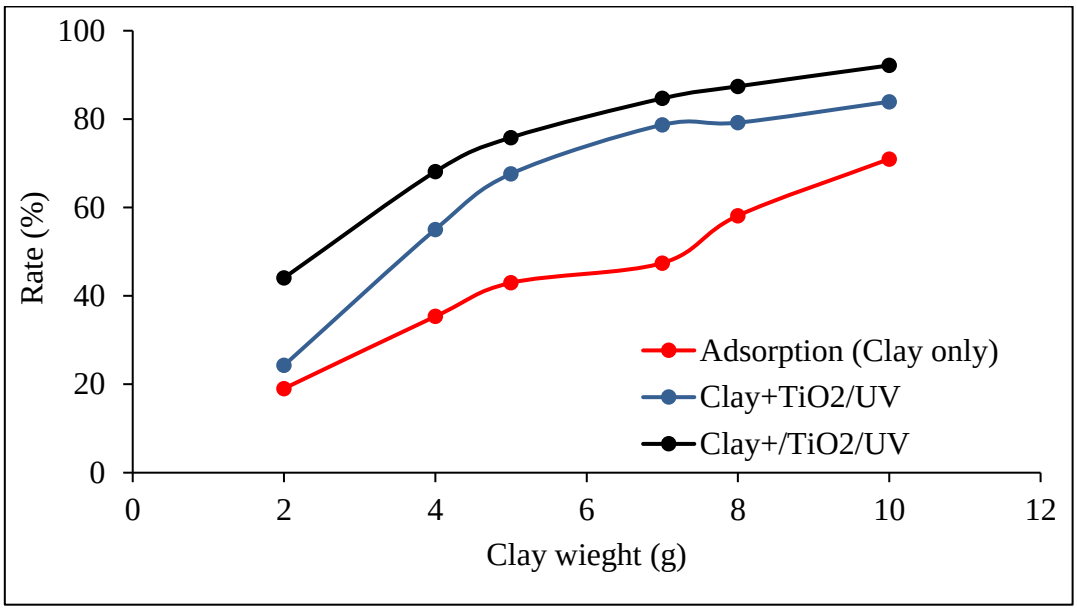


Figure 9. Effect of mass on safranin degradation rate
C_0 = 7 mg/L, T = 25 °C

3*.2.2.2. Effect of initial concentration*

Figure 10 presents the influence of the initial concentration on the degradation kinetics of the dye in the presence of each composite material. In each case, there is a decrease in the rate of degradation of the molecule with increasing concentration. The highest degradation percentages were obtained for a concentration of 1 mg/L, i.e., 81 % and 93 % respectively with the clay+TiO_2 and clay/TiO_2 composite. These results could be explained by the fact that increasing the initial concentration leads

to a decrease in the transparency of the solution, which limits the ability of light to interact with the safranin molecule (Sun *et al.*, 2015). . These results are in agreement with those of Reza *et al.* (2019), who also observed a decrease in the degradation rate as a function of initial concentration during the photocatalytic degradation of violet-3B dye.

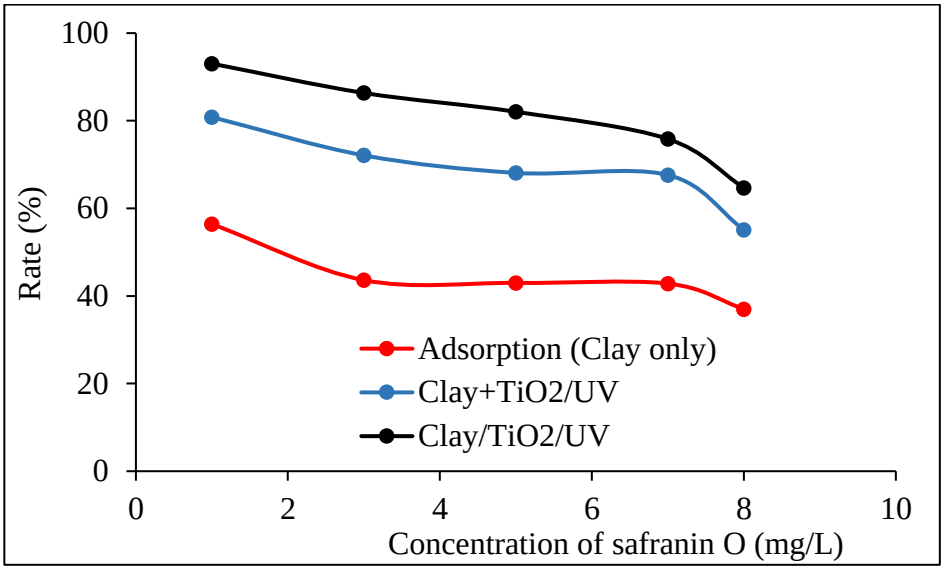


Figure 10. Effect of initial concentration on degradation rate
safranin; m = 5 g, T = 25 °C

3.2.2. Comparison of safranin elimination kinetics by the two processes

The comparison of the adsorption and degradation kinetics presented in **Figure 11** reveals an improvement in the dye elimination process by photocatalysis in the presence of the two types of composites compared to the adsorption process.

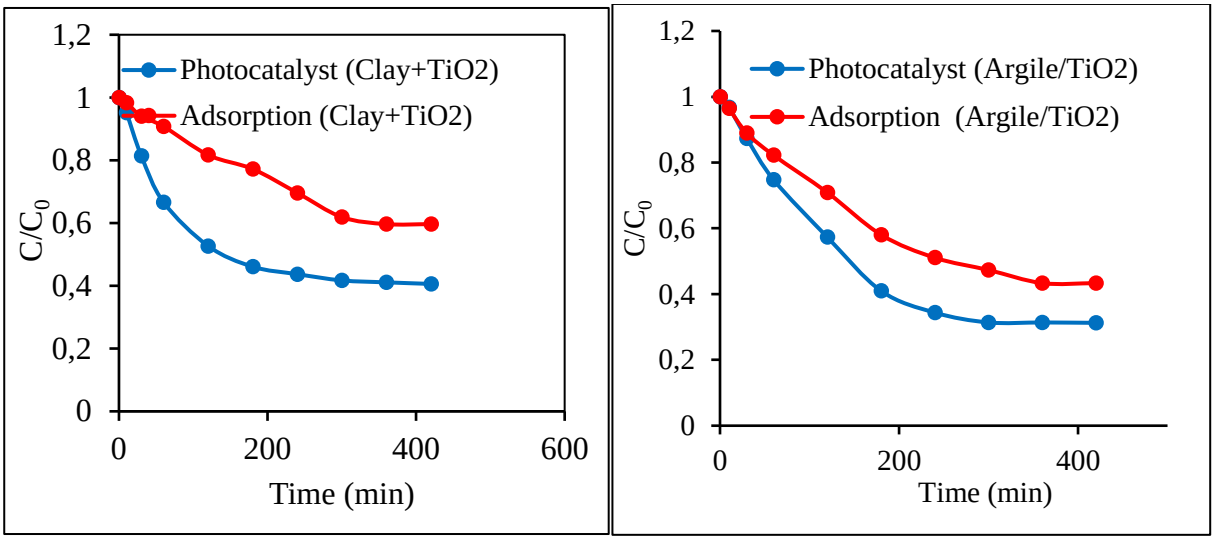


Figure 11. Comparison of safranin removal rates by adsorption and by
photocatalysis; Co = 7 mg/L; m = 5g

The results showed dye degradation rates of 68 % and 76 % respectively with the Clay+TiO_2 and Clay/TiO_2 composites. On the other hand, with the adsorption process, elimination rates of 40 % and

53 % were obtained respectively with the same composites (Clay+TiO_2 and Clay/TiO_2). These observations demonstrate the effectiveness of the photocatalytic process in the degradation of safranin due to the joint action of clay and TiO_2. These results highlight the photocatalytic activity of the Clay+TiO_2 and Clay/TiO_2 composites.

Conclusion

The use of hybrid beads composed of clay and TiO_2 shows promising potential for the removal of organic dyes, such as safranin. The experiments demonstrated that these hybrid beads were able to efficiently adsorb and degrade the dye, with significant removal rates during photocatalysis. The adsorption of the dye onto the clay beads was observed, showing that these porous adsorbents were able to retain the dye due to their high adsorption capacity. The addition of TiO_2 to the clay beads improved the photocatalytic performance, allowing faster degradation of the dye. The clay/TiO_2 hybrid beads showed higher removal rates than the clay+TiO_2 beads, indicating that the combination of the two materials had a synergistic effect on dye removal. It emerges from this study that the clay+TiO_2 and clay/TiO_2 materials can therefore be considered as catalysts with high adsorption capacity, capable of degrading a pollutant under the effect of UV light. Further research will be necessary to optimize the composition and properties of these hybrid materials.

Acknowledgement. The technical inputs of Mr TRAORE Karim Sory of Sciences Environment laboratory are acknowledged.
Disclosure statement: *Conflict of Interest:* The authors declare that there are no conflicts of interest.
Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

References

Aaddouz M., Azzaoui K., Akartasse N., *et al.* (2023). Removal of Methylene Blue from aqueous solution by adsorption onto hydroxyapatite nanoparticles, *Journal of Molecular Structure*, 1288, 135807, https://doi.org/10.1016/j.molstruc.2023.135807

Abu Rub H., Deghles A., Hamed O., *et al.* (2023). Cellulose based polyurethane with amino acid functionality: Design, synthesis, computational study and application in wastewater purification, *International Journal of Biological Macromolecules*, 239, 124328, https://doi.org/10.1016/j.ijbiomac.2023.124328

Ahmed, H.R., Radha, F.H.S., Agha, N.N.M. *et al.* (2023) Characterization and evaluation of Moringa Oleifera Leaves Green Powder and its alkali-activated form as eco-friendly biosorbent for the effective removal of safranin dye from synthetic wastewater. *Reac. Kinet. Mech. Cat.*, 136, 2181–2201. https://doi.org/10.1007/s11144-023-02438-4

Albuquerque B. R., Oliveira M. B. P. P., Barros, L., & Ferreira, I. C. F. R. (2021) Could fruits be a reliable source of food colorants? Pros and cons of these natural additives. *Critical Reviews in Food Science and Nutrition*, 61(5), 805-835. https://doi.org/10.1080/10408398.2020.1746904

Asfaram A., Ghaedi M., Hajati, S., Goudarzi A., & Dil E. A. (2017) Screening and optimization of highly effective ultrasound-assisted simultaneous adsorption of cationic dyes onto Mn-doped Fe3O4-nanoparticle-loaded activated carbon. *Ultrasonics Sonochemistry*, 34, 1-12. https://doi.org/10.1016/j.ultsonch.2016.05.011

Chong M. N., Jin, B., Chow C. W. & Saint C. (2010) Recent developments in photocatalytic water treatment technology : A review. *Water research*, *44*(10), 2997-3027.

Dalli M., Azizi, S., Kandsi F., Gseyra N. (2021) Evaluation of the in vitro antioxidant activity of different extracts of Nigella sativa L. seeds, and the quantification of their bioactive compounds. *Materials Today: Proceedings*, 45, 7259-7263. https://doi.org/10.1016/j.matpr.2020.12.743

Elsherif K. M., El-Dali A., Ewlad-Ahmed A. M., Treban A., Alttayib I. (2021), Removal of Safranin Dye from Aqueous Solution by Adsorption onto Olive Leaves Powder, *J. Mater. Environ. Sci.*, 12(3), 418-430

García-Guzmán P., Medina-Torres L., Josefa Bernad-Bernad Ma., Calderas F., Manero O. (2023) Study of the cholesterol adsorption and characterization of montmorillonite and bentonite clay. *Materials Today Communications*, *35*, 105604. doi.org/10.1016/j.mtcomm.2023.105604

Garg D., Majumder C. B., Kumar S. & Sarkar B. (2019) Removal of Direct Blue-86 dye from aqueous solution using alginate encapsulated activated carbon (PnsAC-alginate) prepared from waste peanut shell. *Journal of Environmental Chemical Engineering*, 7(5), 103365. https://doi.org/10.1016/j.jece.2019.103365

Gaya U. I. & Abdullah A. H. (2008) Heterogeneous photocatalytic degradation of organic contaminants over titanium dioxide : A review of fundamentals, progress and problems. *Journal of Photochemistry and Photobiology C: Photochemistry Reviews*, 9(1), 1-12. https://doi.org/10.1016/j.jphotochemrev.2007.12.003

Jodeh, S., Hamed, O., Melhem, A. *et al.* (2018) Magnetic nanocellulose from olive industry solid waste for the effective removal of methylene blue from wastewater. *Environ. Sci. Pollut. Res.*, 25, 22060–22074. https://doi.org/10.1007/s11356-018-2107-y

Krępska M., Lasoń-Rydel M. & Jagie J. (2016) Charakterystyka, w\laściwości, perspektywy i trudności stosowania niebieskich barwników naturalnych do barwienia produktów spożywczych. *Technologia i Jakość Wyrobów*, 61.

Krishna Moorthy A., Govindarajan Rathi B., Shukla S. P., Kumar K. & Shree Bharti V. (2021) Acute toxicity of textile dye Methylene blue on growth and metabolism of selected freshwater microalgae. *Environmental Toxicology and Pharmacology*, 82, 103552. https://doi.org/10.1016/j.etap.2020.103552

Liu T., Aniagor C. O., Ejimofor M. I., Menkiti M. C., Wakawa Y. M., Li J., Akbour R. A., Yap P.-S., Lau S. Y.& Jeevanandam, J. (2023) Recent developments in the utilization of modified graphene oxide to adsorb dyes from water : A review. *Journal of Industrial and Engineering Chemistry*, 117, 21-37. https://doi.org/10.1016/j.jiec.2022.10.008

Manova E., Aranda P., Angeles Martín-Luengo M., Letaïef, S. & Ruiz-Hitzky E. (2010) New titania-clay nanostructured porous materials. *Microporous and Mesoporous Materials*, 131(1-3), 252-260. https://doi.org/10.1016/j.micromeso.2009.12.031

Mohan D., Singh K. P. & Singh V. K. (2008) Wastewater treatment using low cost activated carbons derived from agricultural byproducts—A case study. *Journal of Hazardous materials*, *152*(3), 1045-1053.

Naghipour D., Taghavi K., Hasanzadeh D., *et al.* (2023). Remove safranin dye from the aqueous solution using ZnO stabilized on zeolite in the presence of ultraviolet light. *Desalination and Water Treatment*, 304, 238-246. https://doi.org/10.5004/dwt.2023.29803

Peramune, D., Manatunga, D. C., Dassanayake, R. S., Premalal, V., Liyanage, R. N., Gunathilake, C., & Abidi, N. (2022). Recent advances in biopolymer-based advanced oxidation processes for dye removal applications : A review. *Environmental Research*, *215*, 114242. https://doi.org/10.1016/j.envres.2022.114242

Tkaczyk A., Mitrowska K. & Posyniak A. (2020) Synthetic organic dyes as contaminants of the aquatic environment and their implications for ecosystems: A review. *Science of The Total Environment*, *717*, 137222. https://doi.org/10.1016/j.scitotenv.2020.137222

Yusuf M., Shabbir M. & Mohammad F. (2017) Natural Colorants : Historical, Processing and Sustainable Prospects. *Natural Products and Bioprospecting*, *7*(1), 123-145. https://doi.org/10.1007/s13659-017-0119-9