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Preparation of Oil Palm Leaves Ash-Supported Titania for the Elimination of Safranin-O Dye in Water

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ABSTRACT. The objective of this study was to develop an approach for incorporating titanium oxide or titania into oil palm leaves ash (OPLA) using a simple procedure. The study process comprised the mixing of oil palm leaves (OPL) powder and titanium tetraisopropoxide in chloroform solvent, followed by the elimination of the solvent to obtain a solid residue. Subsequently, the residue obtained was calcined at 500 °C for 5 hours, leading to the production of a yellow-light powder. The results of product characterization using X-ray diffraction (XRD) and scanning electron microscopy analysis showed the presence of titanium oxide in OPLA. In addition, the wide diffractogram detected in XRD analysis revealed the presence of a silica peak. The un-sharp peaks in the regions of 25.6°, 38.2°, 47.9°, 54.5°, and 62.9° showed that the TiO_2 particles were pure anatase, and no peaks of other TiO_2-anatase phases were detected. Analysis using SEM showed that the surface of the material obtained was irregular and tended to have a hollow shape, while energy dispersive X-ray analysis revealed a SiO_2 content of approximately 73%. However, the titanium element or titania was not detected possibly due to its small concentration. The material obtained also had a good catalytic activity for safranin-O dye elimination under sunlight irradiation, which served as the activation energy source. Based on these findings, the use of OPLA (a side product of oil palm plantations) could be evaluated economically due to its effective role in the catalytic process despite the addition of a small amount of titania catalyst.

Keywords: Oil palm leaves ash, titania, safranin-O, sunlight irradiation.

INTRODUCTION

Waste biomaterials are substances with the potential to be utilized as attractive supporting materials due to their widespread availability, non-toxic nature, and cost-effectiveness. In addition, biomasses or their derivative compounds have similar properties and can facilitate the dispersion of metal and/or metal oxide particles, leading to enhanced stability and recoverability of small particles. Certain biomasses are widely recognized as silica sources (Vaibhav et al., 2015; Permatasari et al., 2016; Patel et al., 2017; Rovani et al., 2018; Sapave et al., 2018; Nasdiyanto et al., 2020), and have been used as supporting materials for metals or metal oxides, including in the standard preparation of TiO_2–SiO_2 composites using commercially available SiO_2 precursors (Pierpaoli et al., 2019). For example, silica obtained from rice husk ash and rice straw was used in the production of TiO_2–SiO_2 composites as photocatalysts for dye discoloration (Yang et al., 2016; Choi et al., 2016).

Recent studies have shown that incorporating nanoscale TiO_2 into black rice husks can enhance the compressive strength of rice husk mortars (Noorvand et al., 2013). In addition, there has been significant progress in this field of study, particularly regarding the use of alternative biomasses as silica sources in ceramic materials preparation (Hubadillah et al., 2018; Hossain et al., 2018; Danewalia et al., 2018; Singh et al., 2020). Among the limited agricultural resources containing silica in their biomasses, oil palm has been reported to be the most demanded. Oil palm leaves (OPL) are widely recognized for their rich silica precursor content, which can be used for various purposes (Onoja et al., 2017; Onoja et al., 2018). In response to the imperative for advancing materials exploration through the use of diverse local natural resources, several preliminary investigations were carried out using OPL as silica precursors. These investigations have yielded materials with unique properties and showed significant potential for fundamental studies (Yudha S et al., 2020; Yudha S et al., 2021). Several studies have also explored the use of oil palm waste, particularly charcoal derived from oil palm shells as supporting materials for a nickel catalyst in the cracking of methyl ester from used cooking oil (Nazarudin et al., 2022).

At present, various reports have explored different composites comprising TiO_2 and supporting materials, primarily focusing on catalytic applications for contaminant degradation in aqueous environments. For example, TiO_2-zeolite and TiO_2-silica composites have been developed as catalysts for the degradation of diverse pollutants, such as safranin-O (SO) (Dong et al., 2012; Ekka et al., 2016). In addition, TiO_2-zeolite composites showed efficacy in degrading SO, methylene blue (MB), and 2,4-dinitroaniline (DNA) within a photocatalytic system. Adsorption on the photocatalyst surface represents a crucial step in the degradation process, with kinetic studies showing that the adsorption of SO conforms to the Langmuir equation (Faghihian & Bahranifard, 2011). Zeolite-supported TiO_2 also showed effectiveness in the degradation of red safranin within a fixed-bed reactor designed for water treatment (Rismanchian et al., 2015). Therefore, the objective of this study was to fabricate titania incorporated into oil palm leaf ash (OPLA) by using a solid-state reaction at high temperatures (500 °C). The samples obtained were then analyzed using scanning electron microscopy, energy dispersive X-ray spectroscopy, X-ray diffraction, and Fourier transform infrared spectroscopy. The catalytic behaviors of the samples for degradation of SO under solar light irradiation experiments were also assessed.

EXPERIMENTAL SECTION

Materials

The materials used comprised titanium tetra-isopropoxide (TTIP) from Sigma Aldrich, as well as hydrochloric acid, nitric acid, and sodium hydroxide from Merck. In addition, the biomass of OPL was obtained from an oil palm plantation farmer, and all glassware was rinsed with an aqua regia solution.

Procedure

After cleaning the OPL with distilled water, the samples were dried and crushed with a juice mixer. A total of 3 g of OPL powder was placed in a crucible, followed by the addition of 50 mL chloroform and stirring using a magnetic stirrer. The reaction mixture was allowed to run for 30 minutes while being stirred to ensure that the OPL powder was completely dissolved in the solvent. While the mixture was still being stirred, 1 mL of TTIP was added. After stirring for 30 minutes, the mixture was allowed to stand at room temperature overnight. The crucible was wrapped in aluminum foil with small holes to ensure slow evaporation of the chloroform. The crucible was inserted into the furnace, and the heating temperature was adjusted to achieve a temperature rise of 500°C in two hours from room temperature. The reaction is allowed to continue at 500 °C for 5 hours. After that, the cooling process was carried out gradually over a period of 24 hours.

The product of the solid-solid reaction was characterized by a variety of commonly used instruments. X-ray diffraction (XRD Mini-Flex, Rigaku), SEM, and energy-dispersive X-ray (EDX) (JEOL JSM 6510 LA) were used for phase, surface, and elemental analysis, respectively. An FTIR spectrophotometer (Compact FT-IR Alpha 2, Bruker) was used to determine the vibration patterns generated by the resulting material. Experiments were performed on 10 mL of SO solutions (10 ppm) with 2 treatments, namely with and without catalyst. The resulting experiments comprised (a) SO without catalyst under dark condition (22 h) + UV irradiation (λ = 254 nm) (3 h), b) SO without catalyst under dark condition (25 h), (c) SO without catalyst under dark condition (22 h) + UV irradiation (3 h) + sunlight irradiation (1 h); (d) SO with TiO_2-OPLA catalyst under dark condition (26 h); (e) SO with TiO_2-OPLA catalyst under dark condition (22 h) + UV irradiation (3 h), and (f) SO with TiO_2-OPLA catalyst under dark condition (22 h) + UV irradiation (3 h) + sunlight irradiation (1 h), with 10 mg catalyst for each treatment. The absorbance was then measured by a spectrophotometer equipped with Agilent-60 at 519 nm. **Figure 1** presents a full schematic diagram of the synthetic pathway and the property analysis, including its application as a photocatalyst.

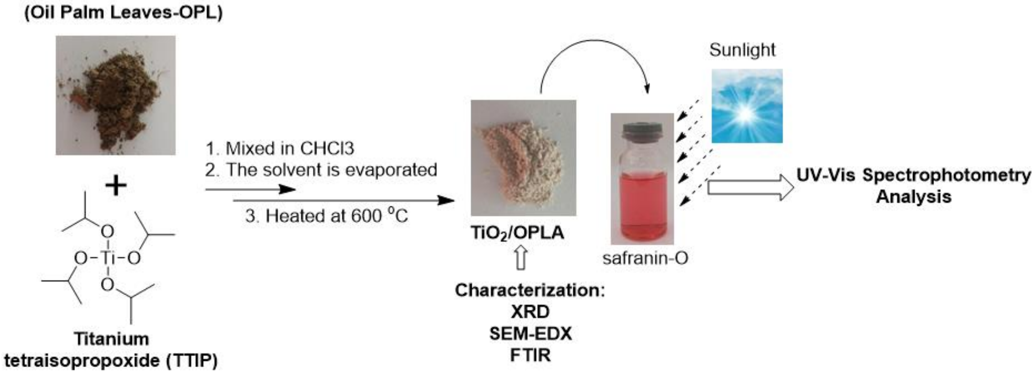


Figure 1. The synthetic processes of TiO_2-OPLA, characterization, and its application

RESULTS AND DISCUSSION

Synthesis of TiO_2-OPLA

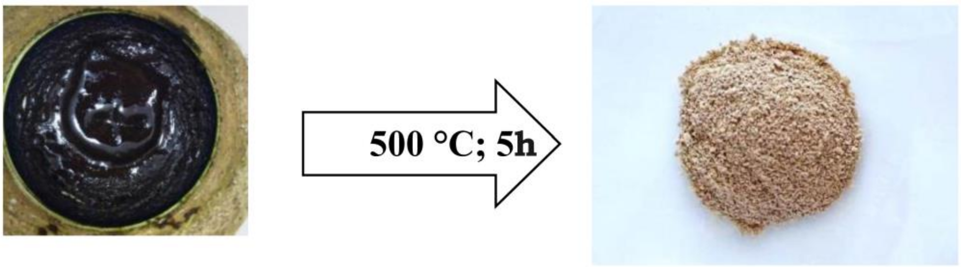
OPL powder in chloroform solvent had a brownish-green color, and the color of the mixture changed to a blackish brown after the addition of TTIP. In addition, the reaction mixture of OPL powder and TTIP was heated at 500 °C for 5 hours to produce a reddish-white powder, as shown in **Figure 2**.

Characterization

The XRD analysis in **Figure 3** showed that when OPL powder was heated with TTIP precursor, amorphous products, such as TiO_2 and OPL ash containing silica (SiO_2) were produced. This was shown by the widening of the silica diffractogram, as well as the absence of sharp TiO_2 diffractogram peaks. Less sharp peaks in the regions of 25.6°, 38.2°, 47.9°, 54.5°, and 62.9° showed that the TiO_2 particles were pure anatase (JCPDS 21-1272). The results of this study also showed that no peaks of other TiO_2 phases were detected. This was consistent with previous studies, establishing that heating the precursor in the open air by the calcination method at a low temperature of 600 °C produced TiO_2 in the anatase phase (Zhai, et al., 1999). In addition, the material could possibly contain compounds or impurities, even in trace amounts, because the experiment was performed on untreated OPL. This was evident through the presence of unidentified small peaks in the XRD.

SEM analysis (**Figures 4a** and **4b**) showed that the product was generally in the form of small powders in the shape of hollow materials. EDX analysis (**Figure 4c**) showed the presence of significant amounts of silica (73.2%) in the indicated area but no titanium oxide. This could be caused by the trace amount of TiO_2 trapped in the SiO_2 matrix, but the XRD peaks were sufficient to show that the resulting material contained TiO_2 (Hong, et al., 2003).

The FTIR analysis of the resulting material revealed the presence of SiO_2, as shown in **Figure 5**. The SiO_2 was indicated by the absorption peak at 1108 cm^{-1}, which showed the presence of a specific Si-O-Si bond. The material also contained a Ti-O-Si interaction due to a peak at 805 cm^{-1}. This finding was consistent with previous studies, reporting that pure SiO_2 did not show a peak in the region, and other reports showed the presence of Ti-O-Si bonds in the 800–1200 cm^{-1} range (Huang et al., 2012). In addition, the wide peak in the 3422 cm^{-1} regions revealed that the material was easily absorbed by moisture due to its correlation with the stretching vibrations of hydroxyl groups and water molecules. The peak was also supported by the presence of a second peak at 1630 cm^{-1}, which revealed the bending vibration of the same molecule.



(a) (b)

Figure 2. Physical change of the reaction mixture (**a**) before (**b**) after calcination

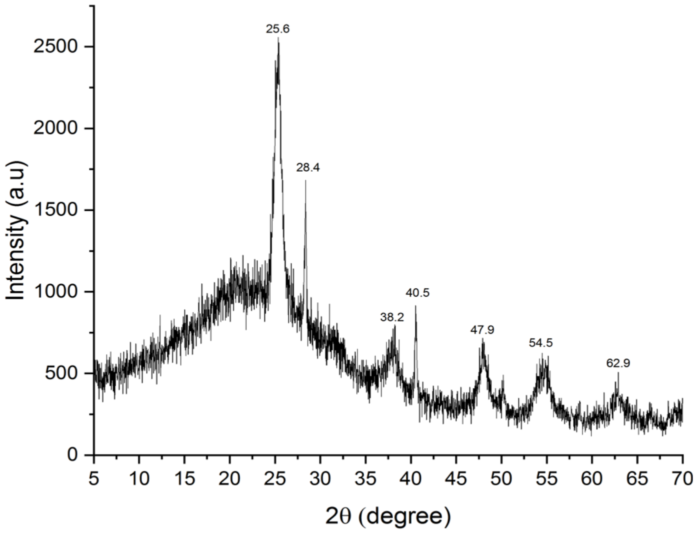


Figure 3. XRD pattern of TiO_2-OPLA

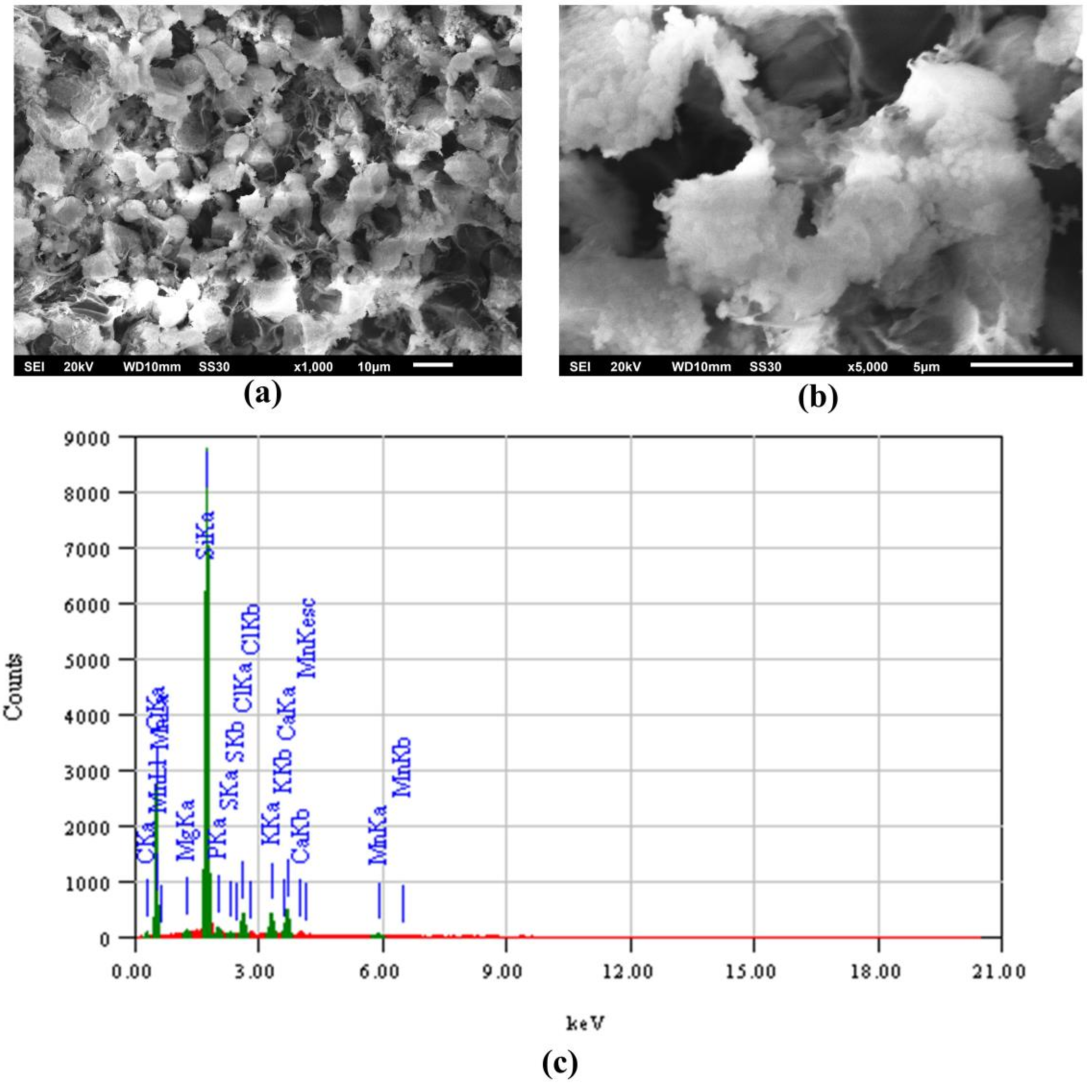


Figure 4. SEM pattern of TiO_2-OPLA (a) 1000×; (b) 5000× and (c) EDX pattern of TiO_2-OPLA

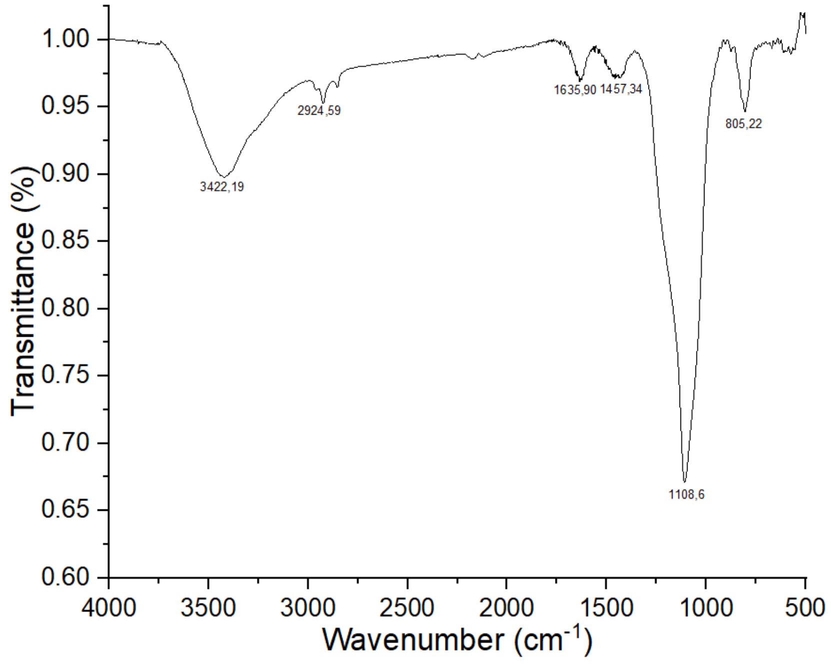


Figure 5. FTIR spectrum of TiO_2-OPLA

Catalytic Applications

Safranin-O (SO) is a dye derived from phenazine, which poses a significant threat to aquatic life when present in wastewater (Jyoti et al., 2016). This textile dye not only raises environmental concerns but also serves as a good candidate for photocatalytic studies due to its high stability to visible light (Sydorchuk et al., 2009). In this study, the elimination of SO in wastewater was explored using TiO_2-OPLA as a catalyst under UV and sunlight irradiation. The experimental procedures started with the preparation of multiple concentrations of SO solution, including 3, 6, 9, 12, and 15 ppm, followed by measurement of their absorbance with a UV-Vis spectrophotometer. Subsequently, these concentrations were plotted to produce a linear regression equation based on the absorbance values obtained. The linear regression equation was then used to determine the change in the concentration of the solutions that had been subjected to treatment with a catalyst and irradiation. The results of the UV-Vis spectrophotometry analysis of SO samples given different treatments are presented in **Figure 6**.

Based on the results, there was no significant difference between the solutions treated without the TiO_2-OPLA catalyst in dark conditions (25 hours) and those treated using a combination of dark conditions and sunlight irradiation (22 hours and 3 hours), as shown in **Figures 6a** and **6b**. This finding suggested that the stability of SO remained unaffected under sunlight irradiation. However, SO without catalyst under dark conditions (22 h), UV irradiation (3 h), and sunlight irradiation (1 h) experienced a decrease in absorbance, leading to an elimination efficiency of 0.56% (**Figure 6c**). Although UV irradiation induced a photolysis reaction for SO, the yield obtained was relatively low. The results revealed that the addition of 10 mg of TiO_2-OPLA catalyst under dark conditions (26 h) led to a slight decrease in absorbance, with an elimination efficiency of 21% (**Figure 6d**). The findings suggested that the presence of catalyst powder in the solution led to a decrease in absorbance due to the adsorption process. This was consistent with the previous studies, reporting that the initial stage of the heterogeneous catalytic reactions comprised the adsorption of treated molecules on and/or into the catalyst (Védrine, 2017). Exposure to UV light for 3 hours activated the TiO_2-OPLA catalyst, leading to a reduction in absorbance due to the photocatalysis process (**Figure 6e**) (elimination efficiency: 29.5%). Meanwhile, exposure to sunlight for 1 hour caused a significant decrease in absorbance and an elimination efficiency of 56%, as shown in **Figure 6f**. The results revealed that the TiO_2 catalyst played an important role in accelerating the elimination of SO compounds in the presence of sunlight irradiation compared to UV irradiation. This observation was in line with previous studies, where the degradation of methylene blue and congo-red was more effective in the presence of TiO_2 under sunlight compared to UV light due to the fast recombination of electrons and holes, which reduced the elimination rate (Bharati et al., 2017). Based on the analysis results in **Figure 6**, sunlight had a substantial influence on increasing the catalytic performance of the catalyst. Consequently, subsequent experiments focused on the use of sunlight as an inducer in SO photocatalysis and as an energy source for the activation of TiO_2-OPLA in the photocatalytic elimination of SO. The UV-Vis spectrophotometry analysis findings are presented in **Figure 7**.

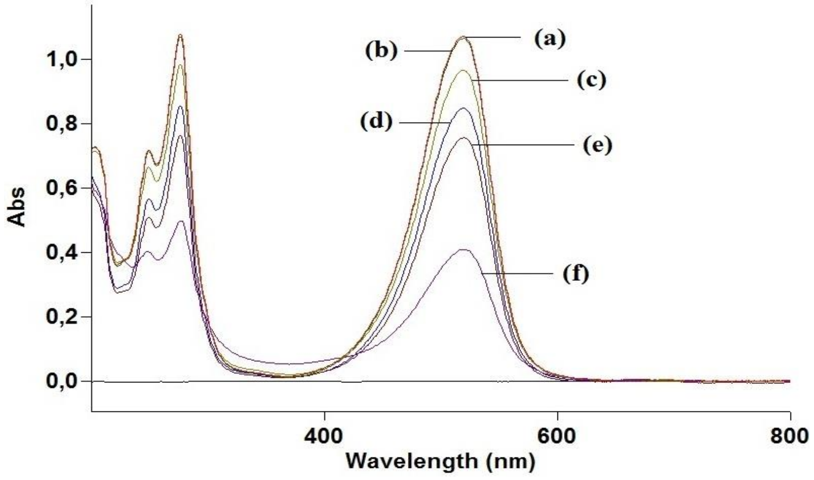


Figure 6. UV–visible spectra of decomposition of SO dye under some reaction conditions: (a) SO without catalyst under dark condition (22 h) + UV irradiation (3 h); (b) SO without catalyst under dark condition (25 h); (c) SO without catalyst under dark condition (22 h) + UV irradiation (3 h) + sunlight irradiation (1 h); (d) SO with TiO_2-OPLA catalyst under dark condition (26 h); (e) SO with TiO_2-OPLA catalyst under dark condition (22 h) + UV irradiation (3 h); (f) SO with TiO_2-OPLA catalyst under dark condition (22 h) + UV irradiation (3 h) + sunlight irradiation (1 h).

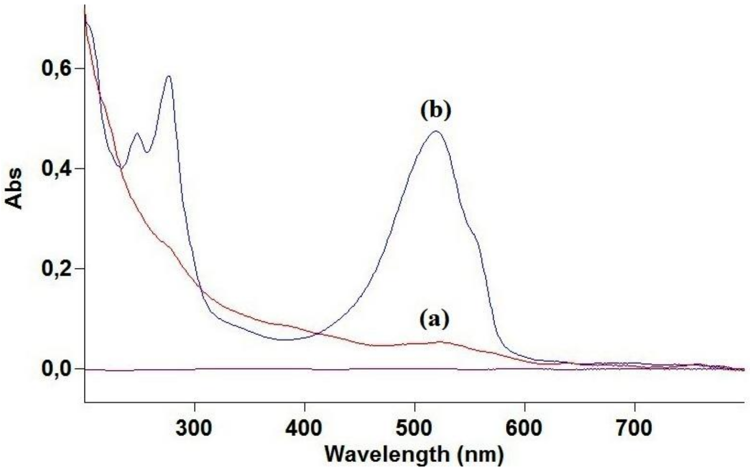


Figure 7. UV–visible spectra of decomposition of SO dye under two reaction conditions: (**a**) SO TiO_2-OPLA catalyst under sunlight irradiation (2 h); (**b**) SO without any catalyst under sunlight irradiation (2 h).

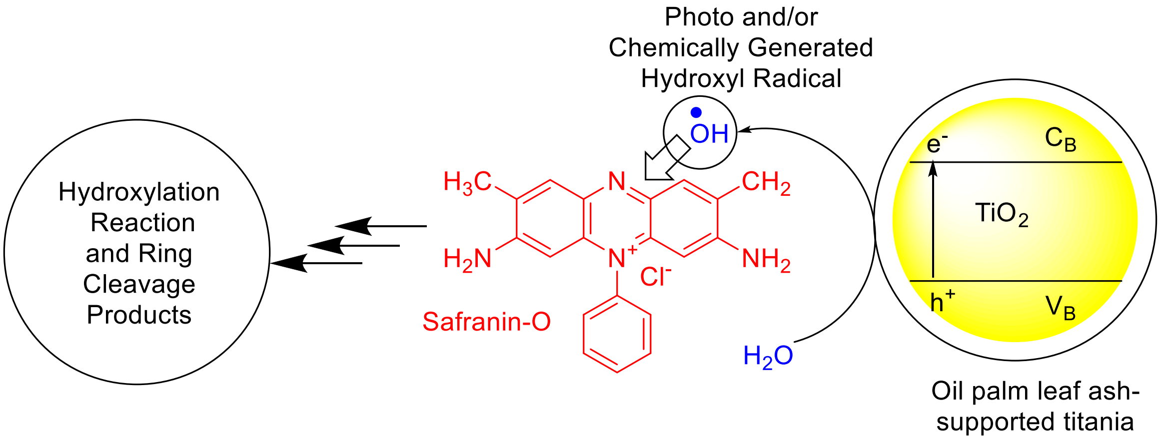


Figure 8. Plausible Reaction Mechanism for Degradation of SO by TiO_2-OPLA

The SO solution treated with the TiO_2-OPLA catalyst exhibited nearly complete elimination (**Figure 7a**). Meanwhile, the untreated solution exposed to sunlight for 2 hours showed photolysis with an elimination efficiency of 55.8% (**Figure 7b**). The synergistic interaction between TiO_2-OPLA particles (which contain a large quantity of SiO_2) and sunlight indicated the potential of OPL waste to be used as a precursor for the production of active catalyst support materials. However, this study predominantly focused on the exploration of catalyst activity in degrading SO compounds without exploring the kinetics and analysis of equilibrium isotherms. From a technical and economic standpoint, the use of sunlight was more profitable due to its constant availability and the non-use of expensive equipment. Several catalytic mechanisms through which TiO_2-SiO_2 composites degraded some essential chemicals have been thoroughly described (Yang et al., 2011; Musaev et al., 2019; Salgado et al., 2019; Panwar et al., 2016). These mechanisms could be used to explain SO elimination by TiO_2-OPLA because OPLA contained a large amount of SiO_2, which produced a plausible mechanism for the current photocatalytic reaction, as shown in **Figure 8**.

Figure 8 shows that the presence of sunlight irradiation on the surface of titania particles causes the excitation of electrons and leaves holes. The process of electron excitation and the presence of holes could cause reactions that produce •OH radicals in water. These radicals broke down the SO compound by starting hydroxylation reactions and breaking the ring, which made the compound break down into simpler, less harmful compounds.

CONCLUSIONS

In conclusion, the use of OPL as a natural source of silica and the incorporation of titanium oxide could be easily fabricated by a solid-state reaction at 500 °C. The resulting material showed that titanium oxide (as anatase) was not completely crystalline compared to silica, which was generally amorphous. TiO_2-OPLA was a material with an irregular shape with good catalytic activity against the elimination of SO when tested outdoors (under sunlight irradiation). The good degradation process reached 56% when SO was treated with the TiO_2-OPLA catalyst under dark conditions, followed by UV and sunlight irradiation for 22 h, 3 h, and 1 h, respectively. Meanwhile, the maximum yield for the degradation process was sufficient in the presence of TiO_2-SiO_2 under sunlight irradiation, which almost degraded safranin-O.

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REFERENCES

Bharati, B., Sonkar, A. K., Singh, N., Dash, D., & Rath, C. (2017). Enhanced photocatalytic degradation of dyes under sunlight using biocompatible TiO_2 nanoparticles. *Materials Research Express*, *4*, 085503.

Choi, C., Hwang, K. J., Kim, Y. J., Kim, G., Park, J. Y., & Jin, S. (2016). Rice-straw-derived hybrid TiO_2–SiO_2 structures with enhanced photocatalytic properties for removal of hazardous dye in aqueous solutions. *Nano Energy*, *20*, 76–83.

Danewalia, S. S., Sharma, G., Thakur, S., & Singh, K. (2016). Agricultural wastes as a resource of raw materials for developing low-dielectric glass-ceramics. *Scientific Reports*, *6*, 24617.

Dong, W., Sun, Y., Ma, Q., Zhu, L., Hua, W., Lu, X., Zhuang, G., Zhang, S., Guo, Z., & Zhao, D. (2012). Excellent photocatalytic degradation activities of ordered mesoporous anatase TiO_2-SiO_2 nanocomposites to various organic contaminants. *Journal of Hazardous Materials*, *229–230*, 307–320.

Ekka, B., Sahu, M. K., Patel, R. K., & Dash, P. (2016). Titania coated silica nanocomposite prepared via encapsulation method for the degradation of Safranin-O dye from aqueous solution: Optimization using statistical design. *Water Resources and Industry*, 22.

Faghihian, H., & Bahranifard, A. (2011). Application of TiO_2–zeolite as photocatalyst for photodegradation of some organic pollutants. *Iranian Journal of Catalysis*, *1*, 45–50.

Hong, S. S., Lee, M. S., Park., S. S., & Lee, G. D. (2003). Synthesis of nanosized TiO_2/SiO_2 particles in the microemulsion and their photocatalytic activity on the decomposition of p-nitrophenol. *Catalysis Today*, *87*, 99–105.

Hossain, S. K. S., Mathur, L., & Roy, P. K. (2018). Rice husk/rice husk ash as an alternative source of silica in ceramics: A review. *Journal of Asian Ceramic Societies*, *6*, 299–313.

Huang, C., Bai, H., Huang, Y., Liu, S., Yen, S., & Tseng, Y. (2012). Synthesis of neutral SiO_2/TiO_2 hydrosol and its application as antireflective self-cleaning thin film. *International Journal of Photoenergy*, *2012*, 620764.

Hubadillah, S. K., Othman, M. H. D., Ismail, A. F., Rahman, M. A., Jaafar, J., Iwamoto, Y., Honda, S., Dzahir, M. I. H. M., & Yusop, M. Z. M. (2018). Fabrication of low cost, green silica based ceramic hollow fibre membrane prepared from waste rice husk for water filtration application. *Ceramics International*, *44*, 10498–10509.

Jyoti, K., & Singh, A. (2016). Green synthesis of nanostructured silver particles and their catalytic application in dye degradation. *Journal of Genetic Engineering and Biotechnology*, *14*, 311–317.

Musaev, K., Mirkhamitova, D., Yarbekov, A., Nurmanov, S., Akbarov, K., & Ruzimuradov, O. (2019). Facile synthesis of SiO_2–TiO_2 photocatalyst nanoparticles for degradation of phenolic water pollutants. *SN Applied Sciences*, *1*, 1164.

Nandiyanto, A. B. D., Ragadhita, R., & Istadi, I. (2020). Techno-economic analysis for the production of silica particles from agricultural wastes. *Moroccan Journal of Chemistry*, *8*, 801–818.

Noorvand, H., Ali, A. A. A., Demirboga, R., Farzadnia, N., & Noorvand, H. (2013). Incorporation of nano TiO_2 in black rice husk ash mortars. *Construction and Building Materials*, *47*, 1350–1361.

Onoja, E., Attan, N., Chandren, S., Razak, F. I. A., Keyon, A. S. A., Mahat, N. A., & Wahab, R. A. (2017). Insights into the physicochemical properties of the Malaysian oil palm leaves as an alternative source of industrial materials and bioenergy. *Malaysian Journal of Fundamental and Applied Sciences*, *13*, 623–631.

Onoja, E., Chandren, S., Ilyana, F., Razak, A., & Wahab, R. A. (2018). Extraction of nanosilica from oil palm leaves and its application as support for lipase immobilization. *Journal of Biotechnology*, *283*, 81–96.

Panwar, K., Jassal, M., & Agrawal, A. K. (2016). TiO_2–SiO_2 Janus particles with highly enhanced photocatalytic activity. *RSC Advances*, *6*, 92754–92764.

Patel, K. G., Shettigar, R.R., & Misra, N. M. (2017). Recent advance in silica production technologies from agricultural waste stream–review. *Journal of Advanced Agricultural Technologies*, *4*, 274-279.

Permatasari, N., Sucahya, T. N., & Nandiyanto, A. B. D. (2016). Review: agricultural wastes as a source of silica material. *Indonesian Journal of Science and Technology*, *1*, 82-106.

Pierpaoli, M., Zheng, X., Bondarenko, V., Fava, G., & Ruello, M. L. (2019). Paving the way for a sustainable and eficient SiO_2/TiO_2 photocatalytic composite. *Environments, 6,* 87.

Rismanchian, M., Barakat, S., Khoshzat, N., Keshavarzi, R., & Mahnaz, S. (2015). Investigation of TiO_2/zeolite photocatalytic activity for Safranin dye removal of aqueous solution. *International Journal of Environmental Health Engineering, 4*(1), 307–320.

Rovani, S., Santos, J. J., Corio, P., & Fungaro, D. A. (2018). Highly pure silica nanoparticles with high adsorption capacity obtained from sugarcane waste ash. *ACS Omega*, *3*, 2618–2627.

Salgado, B. C. B., & Valentini, A. (2019). Evaluation of the photocatalytic activity of SiO_2@TiO_2 hybrid spheres in the degradation of methylene blue and hydroxylation of benzene: kinetic and mechanistic study. *Brazilian Journal of Chemical Engineering*, *36*, 1501–1518.

Sapave, N., & Hanafi, M. F. (2018). Production of silica from agricultural waste. *Archives of Organic and Inorganic Chemical Sciences, 3*(2), 342–343

Singh, J., Boddula, R., & Jirimali, H. D. (2020). Utilization of secondary agricultural products for the preparation of value-added silica materials and their important applications: a review. *Journal of Sol-Gel Science and Technology*, *96*, 15–33.

Sydorchuk, V., Khalameida, S., & Zazhigalov, V. (2009). Mechanochemical synthesis of barium titanate and its photocatalytic properties. *Annales Universitatis Mariae Curie-Sklodowska sectio AA – Chemia*, *64*, 159–168.

Vaibhav, V., Vijayalakshmi, U., & Roopan, S. M. (2015). Agricultural waste as a source for the production of silica nanoparticles. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, *139*, 515–520.

Védrine, J. C. (2017). Heterogeneous Catalysis on Metal Oxides. *Catalysts*, *7*, 341.

Yang, D., Fan, T., Zhou, H., Ding, J., & Zhang, D. (2011). Biogenic hierarchical TiO_2/SiO_2 derived from rice husk and enhanced photocatalytic properties for dye degradation. *Plos One*, *6*, e24788.

Yang, Z. Y., Shen, G. Y., He, Y. P., Liu, X. X., & Yang, S. J. (2016). Preparation of TiO_2/SiO_2 composite oxide and its photocatalytic degradation of rhodamine B. *Journal of Porous Materials*, *23*, 589–599.

Yudha S, S., Falahudin, A., Asdim., & Han, J. I. (2021). In situ preparation of gold–silica particles from a mixture of oil palm leaves and chloroauric acid for reduction of nitroaromatic compounds in water. *Waste and Biomass valorization*, *12*, 3773–3780.

Yudha S, S., Falahudin, A., Kaus, N. H. M., Thongmee, S., Ikram, S., & Asdim. (2020). Preliminary synthesis of calcium silicates using oil palm leaves and eggshells. *Bulletin of Chemical Reaction Engineering & Catalysis*, *15*, 561–567.

Yudha S, S., Robkhob, P., Imbon, T., Falahudin, A., & Asdim. (2020). ZnO-SiO_2 and Zn_2SiO_4 synthesis utilizing oil palm leaves for degradation of methylene blue dye in aqueous solution. *Journal of the Indonesian Chemical Society, 3*, 94–100.

Zhai, J., Zhang, L., & Yao, X. (1999). Effects of composition and temperature on gel-formed TiO_2/SiO_2 films. *Journal of Non-Crystalline Solids*, *260*, 160–163.