Journal of Peat Science and Innovation
Volume 1 Number 1 June 2022, pp1-15

JPSI
Journal of Peat Science and Innovation

Review Article

A Review: Development of Photocatalyst Materials and Its Performance for Humic Acid Removal in Peatwater

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Received 01 January 2022
Revised 01 May 2022
Accepted 01 June 2022

Citation:
Beladona, S.U.M., Putra, R., Alfanaar, R., Sylvani, M.M., Alyatikah, E., Safitri, R., Susanti, I., & Iqbal, R.M. (2022). A Review: Development of Photocatalyst Materials and Its Performance for Humic Acid Removal in Peatwater. *Journal of Peat Science and Innovation*, 1(1), pp1-10.

Abstract. Peat is a wetland region in Indonesia with a very high water storage capacity. However, the amount of humic compounds obtained from the accumulation of organic substances in peat makes the water unable to be used for daily purposes. Peat water treatment with several methods has been carried out, such as coagulation, electrocoagulation, flocculation, and filtration. However, the result of clumping or filtering in such a way raises new environmental problems. One method which effective and efficient used in peat water treatment is photocatalytic. Several materials based-semiconductor was developed as a photocatalyst, another modification of photocatalyst is combining the adsorbent as a porous supporting photocatalyst which can improve its performance.

Keywords: Humic acid, Metal oxides, Peat water, Perovskite, Photocatalytic.

1. Introduction

Indonesia is one country that has the most significant wetland areas in the world. The territory belongs to the wetlands, one of which is called peatland. Indonesia's peatlands potential reaches 14.9 million hectares. Based on research conducted by the Center for Research and Development of Land Resources and Soil Research Institute in 2011, particularly in Palangka Raya in Central Kalimantan province spread of peatlands reached 3.1 million hectares.

Peat is a type of soil formed from plant debris accumulated half-rotted, so the organic matter content is high enough. Depending on the depth and type of peat, the water storage capacity on peat soil is very diverse. Peat at depths of 0-50 and 50-100 cm is classified as hemic peat, which has a moisture content of from 541.82 to 719.41%, while peat at a depth of 100-150 cm is classified as fabric peat and has a water storage capacity of 815.1 to 1020.59%. Peat water has a pH ranging from 3.7 to 5.3, and

turbidity is caused by the presence of substances such as sludge suspended, organic substances, and iron metal, the brown color with 124-850 Pt-Co scale, and smells due to natural decomposition materials (Susandi et al., 2015).

The characteristics of peat water are not in accordance with the water quality standardized by the Department of Health through PERMENKES 32/Ministry of Health/PER/2007, namely pH ranging from 6.5 to 8.5, odorless, the maximum total dissolved solids (TDS) of 1000 mg/L, the maximum turbidity of 25 NTU, tasteless, the maximum temperature of 3 °C, the maximum color of 50 TCU, and the amount of organic matter in water quality that includes $KMnO_4$ must have the maximum level of 10 mg/L, and the maximum of Fe content is 1 mg/L. One with a high percentage of organic matter contained in the peat water is humic acid. Humic acid is an aromatic compound with carbonyl groups and phenolic formed by the degradation of plant and microbial materials. The humic acid content causes the pH of peat water to be low and the color change to be brown (Andayani & Bagyo, 2011). The molecular structure of humic acid is shown in Figure 1. Based on the above explanation, it is necessary to degrade the humic acid compound as one of the ways for peat water treatment, before it is used for daily purposes.

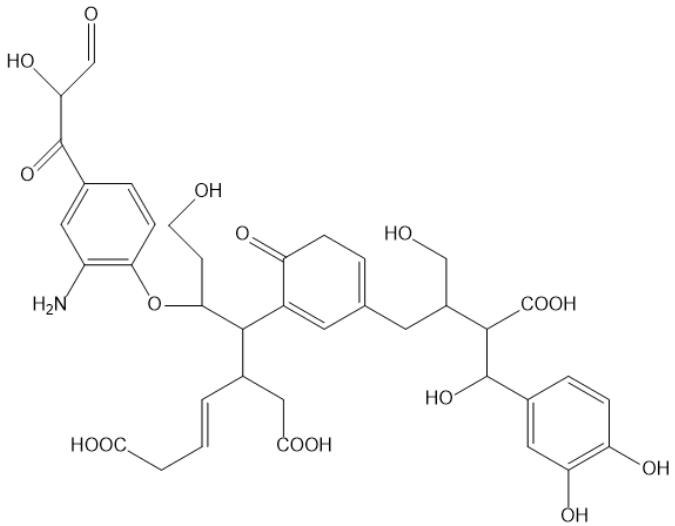

Figure 1. The molecular structure of humic acid

2. Previous Research for Peatwater Treatment

Peat water treatment has been carried out with several methods, such as coagulation, electrocoagulation, flocculation, and filtration. However, these methods only focused on separating organic substances from peat water. In contrast, the rest of these methods' filters or clumping substances are not reprocessed into useful material, giving rise to new environmental problems. The several techniques used for peat water treatment are listed in Table 1.

Table 1. Previous methods for peat water treatment

No.	Peatwater Treatment Methods	Weakness	Reference
1.	Coagulation method with coagulant mixture (Corn Seed, Moringa, and Watermelon)	Require large amounts of seed mixtures because of the relatively small size	(Rehansyah et al., 2007)
2.	Peat Water Treatment Using Chitosan-Silica Composite as Adsorbent	The reaction is not spontaneous and endothermic at low temperatures	(Zulfikar et al., 2014)
3.	Coagulation-Flocculation method Atmosphere Bases	Results precipitation does not reprocess	(Suherman & Sumawijaya, 2013)
4.	Reduction Peat Humic Acid in Water Using Powdered Eggs Without Shells Calculated as Adsorbent Economical	A small surface area material	(Daud et al., 2016)

5.	Electrocoagulation Method with Batch and Continuous Reactor Scheme Using Type Aluminum Electrolytic	Requires high-voltage electricity for desalination of peat in large capacity	(Suwanto et al., 2017)

3. Mechanism of Photocatalytic Degradation

Photocatalytic is a combination of photochemical and catalytic processes that use semiconductor material as a catalyst. The photocatalytic degradation process begins with the formation of electron-hole pairs in the semiconductor particles due to the impulse of photons or electromagnetic waves that cause the electron movement from the valence band to the conduction band. Photocatalyst converts photons into chemical energy, which will produce hydroxyl radicals, and oxidize the organic compounds (pollutants) so that the water will be clean as an inseparable part of contaminants. These pollutants are decomposed into CO_2 and H_2O that are environmentally friendly, as shown in Figure 2 (Sucahya et al., 2016). Through photodegradation, pollutants would be broken down into simpler components. At the same time, the heavy metals in the waste or peat water would be reduced so that the water be free of contaminants. Degradation of organic pollutants such as humic acid by photocatalytic led to the deconstruction of molecular structure that resulted in a change of color (Fatimah et al., 2022). The photocatalytic mechanism is shown in Figure 2.

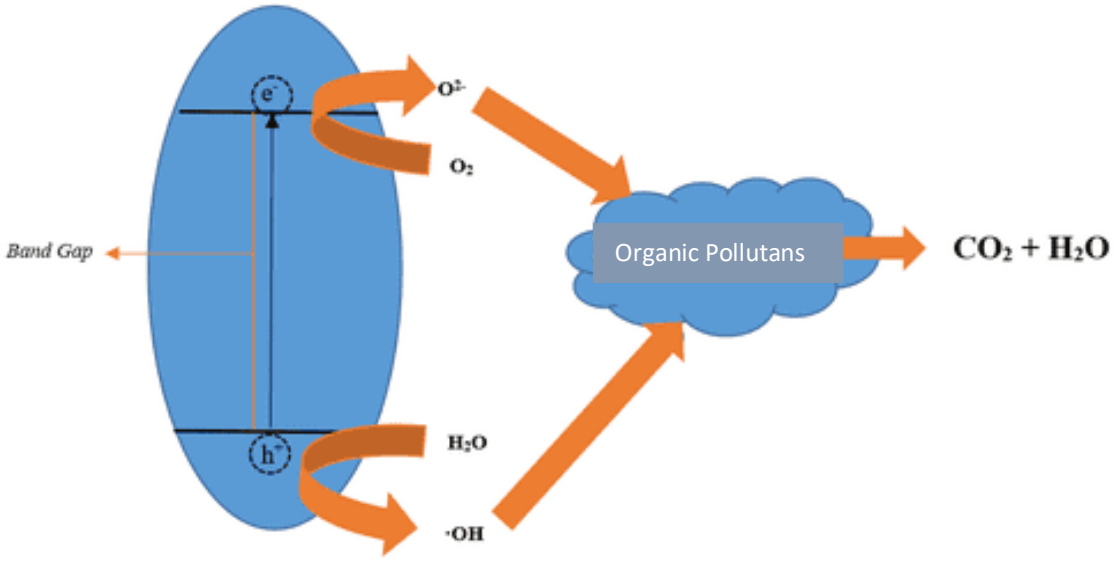

Figure 2. The mechanism of photocatalytic degradation

4. Photocatalyst Materials

The photocatalyst is a catalyst to accelerate the process of photoreaction. If a photocatalyst is subjected to a photon, it will form two kinds of reactive substances such as hydroxyl radicals (•OH) and superanion oxide (O^{2-}) (Kumar et al., 2017). The semiconductor material is a material that can be used as a photocatalyst because it has an empty energy area called the band gap energy which is located between the valence band and the conduction band. The magnitude of the bandgap energy can be measured using a wavelength of light that can excite the electrons (Bey, 2009; Qodri, 2011).

The semiconductor material that absorbs photons must have an energy equal to or greater than the bandgap energy possessed by the photocatalyst, which is sufficient to excite electrons (Qodri, 2011). Electrons (e^-) on the valence band to the conduction band will move and cause the formation of the hole (h^+) in the valence band. Particles (e^-) are formed to initiate the process of oxidation of pollutants, while gap (h^+) plays a role in the oxidation reaction of contaminants on the surface of the photocatalyst material. The pairs of hole-electron will automatically recombine on the semiconductor (Iqbal et al.,

2020), and it will lead to the degradation process. The most well-known material as a photocatalyst is TiO2 (Iqbal et al., 2020; Iqbal et al., 2021; Susanti et al., 2021), but another semiconductor also has potential for photocatalyst application. Table 2 shows some semiconductor material that is widely used by researchers as a photocatalyst to degrade pollutants.

One of the photocatalyst materials which has the lowest bandgap energy value is CuS, as determined by Amurugam and Jagannathan (Amurugam & Jagannathan, 2017). CuS has an energy value of the bandgap of 1.59 eV, while CuS and CusS/CdS (1 wt%) have a band gap of 2.25 and 2.19, respectively. Then modification of photocatalysts with compositing CuS and CdS by the hydrothermal method to form the material CuS/CdS to degrade compounds of organic pollutants. Modifications are made to obtain a more optimal result degradation. The changes in the amount of bandgap energy of the photocatalyst material will directly impact to the results of degradation. The use of the CuS/CdS composite has a great performance compared to CuS and CdS for methylene blue degradation. The formation of mixed semiconductor material leads to the recombination of electron-hole pairs with a higher number.

Table 2. Energy bandgap semiconductor material

No.	Material	Energy Band Gap (eV)	Reference
1.	CuS	1.59	(Amurugam & Jagannathan, 2017)
2.	Cu-S/TiO_2	1.90	(Haris et al., 2014)
3.	Cus/CdS	2.19	(Amurugam & Jagannathan, 2017)
4.	CdS	2.25	(Amurugam & Jagannathan, 2017)
5.	C-ZnO	2.69	(Lavand & Malghe, 2015)
6.	Zn-TiO_2	2.83	(Aware & Jadhav, 2016)
7.	TiO_2	2.96	(Aware & Jadhav, 2016)
8.	ZnO	3.08	(Lavand & Malghe, 2015)
9.	TiO_2	3.22	(Almu'minin, 2015)
10.	Ce-ZnO	3.26	(Birben et al., 2017)
11.	ZrO_2	3.64	(Fakhri et al., 2016)
12.	ZnO_2	3.80	(Wicaksono et al., 2013)
13.	C-ZrO_2	3.81	(Fakhri et al., 2016)

Another study conducted by Fakhri et al. used ZrO_2 material that has a bandgap energy value is 3.64 eV (Fakhri et al., 2016). Then, this material is modified by carbon dopants to form the photocatalyst of C-ZrO_2. Material modifications of ZrO_2 to C-ZrO_2 increased the amount of the bandgap energy from 3.64 to 3.81 eV. However, the C-ZrO_2 can degrade the compound amoxicillin 10% higher than compared with ZrO_2. It is also influenced by the wavelength of irradiation light during the reaction. The band gap energy must be suitable to the wavelength of light to achieve greater results for the degradation of organic pollutants. On the other hand, the researchers must be selective in selecting the light source which is suitable for semiconductor or photocatalyst material.

Other materials that could potentially be used as a photocatalyst also include perovskite. Perovskite is a compound with the empirical formula ABO_3. Based on the empirical formula, a more abundant cation occupies site A and smaller cations occupy position B. Grid compound formed depends on the radius of the cation (A place) and cation (B site) electronegativity (Iqbal et al., 2018; Iqbal et al., 2018; Nurherdiana et al., 2019; Nurherdiana et al., 2017). Perovskite material is widely developed and applied in the photocatalyst field because it is capable of recombining electron-hole, as shown in Table 3.

Table 3. Bandgap energy of perovskite-based photocatalyst material

No.	Material	Energy Band Gap (eV)	Reference
1.	$BiFeO_3$	1.68	(Liu et al., 2017)
2.	$BiFeO_3$-TiO_2	1.77	(Liu et al., 2017)
3.	$LaFeO_3$	2.10	(Kanhere & Chen, 2014)
4.	$LaNiO_3$	2.26	(Li et al., 2010)
5.	$LaCoO_3$	2.70	(Kanhere & Chen, 2014)
6.	$BaTiO_3$	3.20	(Yadav et al., 2018)
7.	$ZnTiO_3$	3.20	(Salavati-Niasari et al., 2016)
8.	$CaTiO_3$	3.50	(Kumar et al., 2018)
9.	$NaTaO_3$	4.00	(Li et al., 2015)

In general, the crystal structure of the perovskite as shown in Figure 3. The connection between the perfect octahedral BO_6 structure will form a cubic lattice. Cation B has a stronger bond with oxygen, while cation A reverse. The crystal structure of perovskite has a corner that connects BO_6 octahedral and 12 cations A coordinated oxygen, which lies between eight octahedral BO_6. Perovskite compounds with the degree of slope BO_6 in different octahedral crystal fields will produce different physical properties. The degree of tilt can affect the band structure, electron and hole transport properties, Photoluminescence, and dielectric behavior (Kanhere & Chen, 2014).

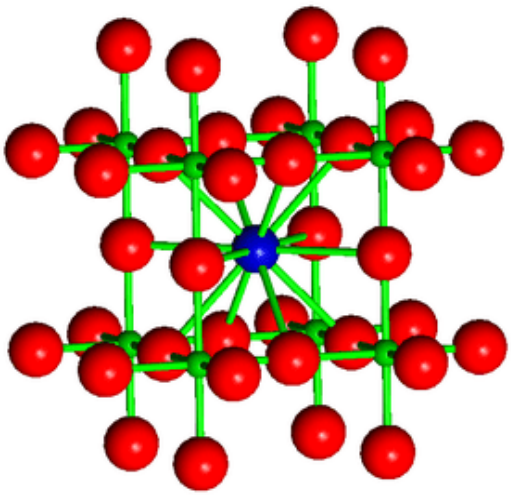

Figure 3. The crystal structure of Perovskite (red: A cation, blue: B cation, green: oxygen) (Kanhere & Chen, 2014)

$BiFeO_3$ is one of the perovskite compounds synthesized by Liu et al. to degrade the compound methyl violet dye (Liu et al., 2017). $BiFeO_3$ synthesized by citrate-nitrate combustion method. The mixture has bandgap energy values relatively low at 1.68 eV, as shown in Figure 4a. $BiFeO_3$ is able to degrade methyl violet under visible light radiation with an efficiency of up to 90%. In this study, the modification to form a composite $BiFeO_3$ photocatalyst and TiO_2 leads to enhancing the photocatalytic activity. $BiFeO_3$-TiO_2 composites have an energy band gap of 1.77 eV, as shown in Figure 4b. Methyl violet degradation efficiency reached 95%, as shown in Figure 5. This material is possible to apply for humic acid degradation as a method for peat water treatments.

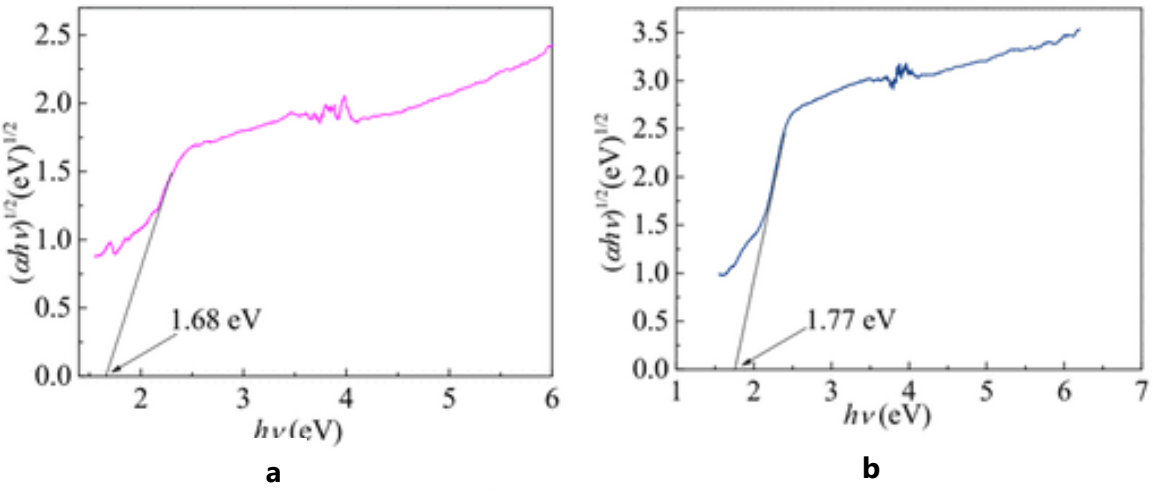

Figure 4. Plot (αhv) 1/2 of the bandgap energy (hv) (a) $BiFeO_3$ and (b) $BiFeO_3$-TiO_2 (Liu et al., 2017)

Another study conducted by Li et al. to degrade phenolic compounds using $NaTaO_3$ compounds were synthesized by the hydrothermal method (Li et al., 2010). $NaTaO_3$ bandgap energy is high at 4.00 eV. These compounds are active under UV radiation, but their modification to provide S-dopant into $NaTaO_3$ improves the photocatalytic performance. S-dopant $NaTaO_3$ was able to degrade phenol with an efficiency of more than 90%, whereas pure $NaTaO_3$ only about 30%. Based on several explanations and the data about photocatalytic performance, all material based-semiconductor is possible to use in the photodegradation process, and also can be used to remove humic acid in peat water or other organic pollutant.

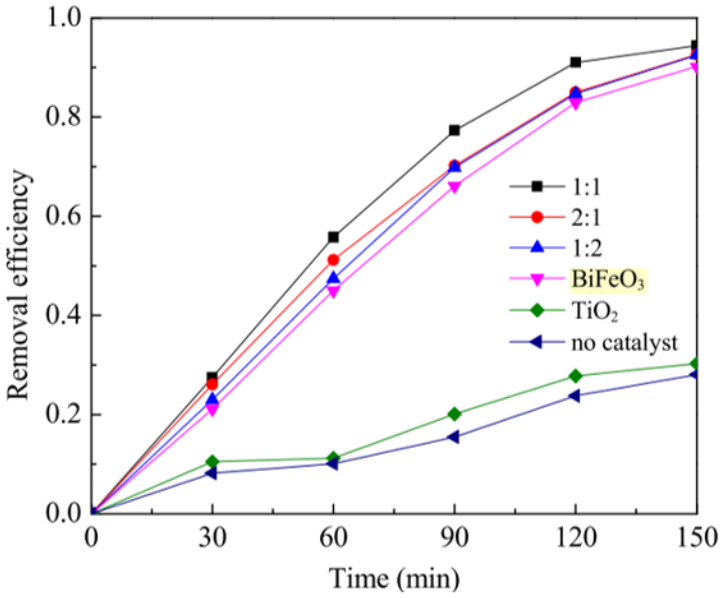

Figure 5. Photocatalytic activity for the degradation of methyl violet under visible light radiation (Liu et al., 2017)

5. Integrated Photocatalyst and Adsorbent

Some materials in the form of metal oxide or perovskite-based semiconductors have been applied as a photocatalyst to degrade pollutants in the water or waste. However, the non-porous surface of the catalyst can significantly affect its photocatalytic. Therefore, researchers carried out various

modifications to the photocatalyst to increase the photocatalytic activity, such as doping material. Composite fabrication is constructed until the merger of photocatalysts and adsorbents.

The adsorbent is a porous material that can be used to absorb pollutants. This material has been widely used by previous researchers to reduce organic and inorganic contaminants in wastewater treatment. Commonly known types of adsorbents such as activated carbon, bentonite, montmorillonite, clay, zeolite, and others.

The use of adsorbents as supporting materials for photocatalyst can increase photocatalytic activity. Pores in adsorbent lead to the increasing specific surface area of the photocatalyst so that the contact between the photocatalyst and pollutants also gets more significant (Iqbal et al., 2021). The number of pollutant particles that are absorbed by the adsorbent and degraded by the photocatalyst can accelerate the photocatalytic process itself. Integrating photocatalyst and adsorbent material is an important step to achieving optimal degradation products. The adsorbent which absorbs pollutant molecules can overcome the problem of the low absorptivity of the photocatalyst material, while the photocatalyst itself acts as an electron-hole recombination agent.

There are various kinds of adsorbents used by previous researchers to serve as photocatalysts supporting materials, such as activated carbon, clay, zeolite, bentonite, montmorillonite, and others. The development of the photocatalyst and the adsorbent material modifications is shown in Table 4.

Peng et al. perform material synthesis of $LaFeO_3$/montmorillonite to degrade Rhodamine B (Peng et al., 2016). The montmorillonite is a type of adsorbent such as clay minerals that are abundantly found with natural morphology such as 2D sheets. In this study, $LaFeO_3$/montmorillonite was synthesized by the sol-gel method. The combination of a perovskite material and adsorbent cause changes in bandgap energy value and specific surface area, as shown in Figure 6a and 6b, respectively. Montmorillonite as a supporting material $LaFeO_3$ led to the increased particular surface area caused by the particle distribution of $LaFeO_3$ on the montmorillonite sheet.

Table 4. Characteristics of integrating the photocatalyst and the adsorbent material

No.	Material	Energy Band Gap (eV)	Specific Surface Area (m^2/g)	Reference
1.	$LaFeO_3$/montmorillonite	2.24	13.15	(Peng et al., 2016)
2.	Fe: ZnO/Zeolite/Fe_3O_4	3.18	82.11	(Kane et al., 2016)
3.	Ag-N-ZnO/Coconut husk-coal-almond (Chac)	2.52	472	(Chen et al., 2017)
4.	Ag-N-ZnO/Coal activated carbon (CAC)	2.55	252	(Chen et al., 2017)
5.	Ag-N-ZnO/Almond activated carbon (AAC)	2.77	219	(Chen et al., 2017)
6.	TiO_2/Bentonite	3.20	18.33	(Surya et al., 2018)
7.	ZnO/Graphene oxide	3.25	26.4	(Durmuz et al., 2019)
8.	TiO_2/Activated carbon	3.45	465	(Peñas-Garzón et al., 2019)
9.	TiO_2/Zeolite	3.10	293	(Liao et al., 2019)
10.	TiO_2/Clay	3.20	116.7	(Hadjltaief, et al., 2019)

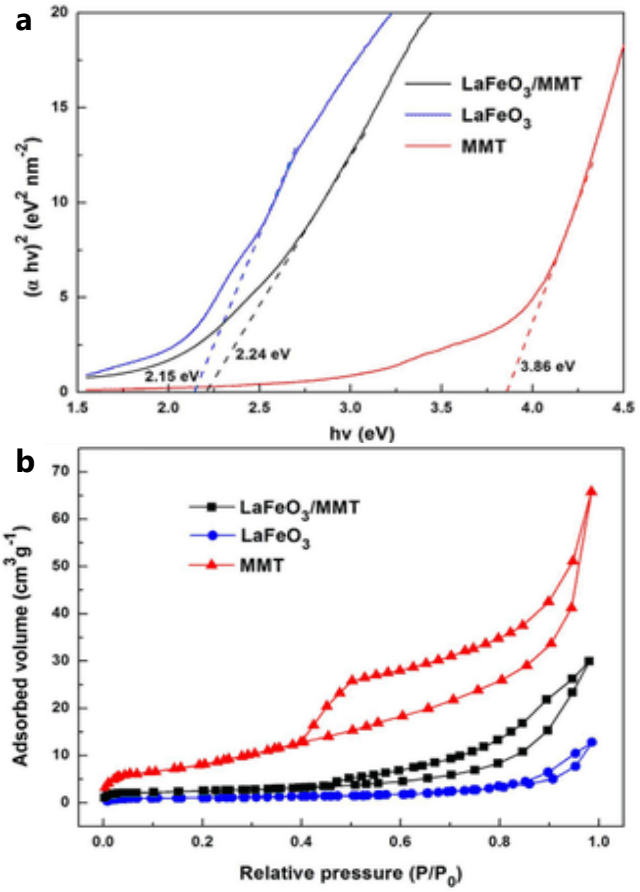

Figure 6. (a) Plot (αhv) 1/2 of the band gap energy (hv) $LaFeO_3$/montmorillonite and (b) N_2 adsorption-desorption isotherms $LaFeO_3$/montmorillonite (Peng et al., 2016)

Chen et al. researched the form of photocatalytic degradation of methyl orange using photocatalyst Ag-N-ZnO and activated carbon support material. Activated carbon used in this study were obtained from various sources such as coconut shell, coal, and almonds (Chen et al., 2017). Activated carbon source variations affect the bandgap energy value and specific surface area material, as shown in Figures 7 and 8.

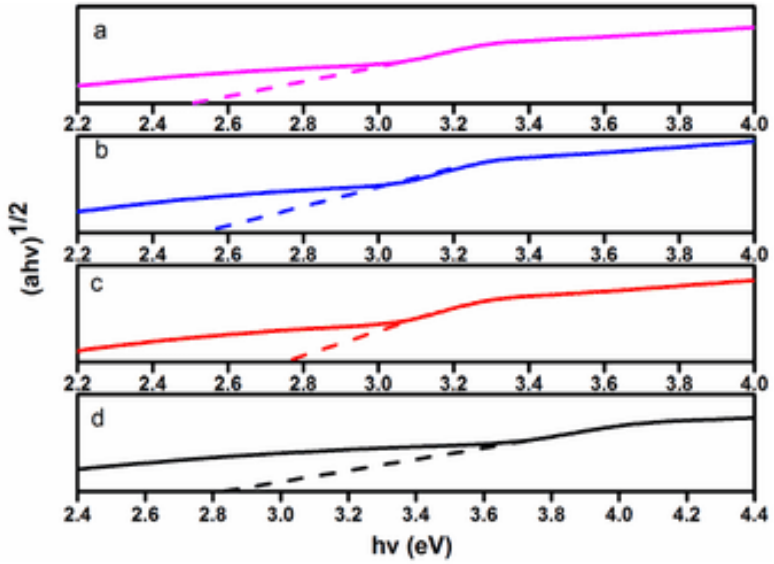

Figure 7. Plot (αhv) 1/2 of the bandgap energy (HV) (a) Ag-N-ZnO/CHAC, (b) Ag-N-ZnO/CAC, (c) Ag-N-ZnO/AAC and (d) Ag-N-ZnO (Chen et al., 2017)

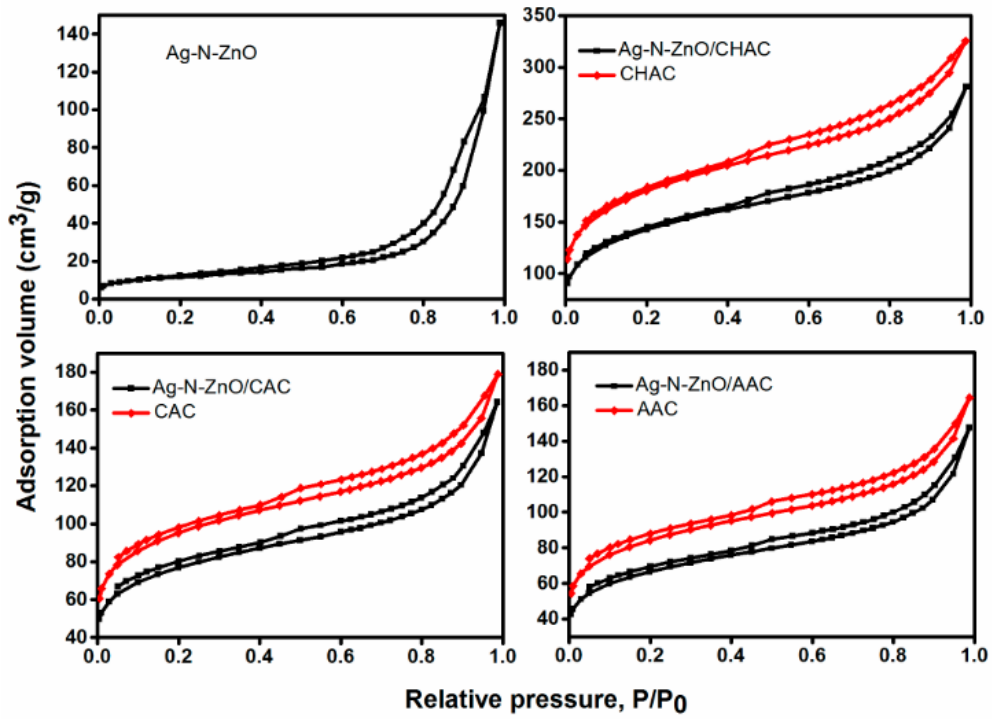

Figure 8. N_2 adsorption-desorption isotherms Ag-N-ZnO/activated carbon (Chen et al., 2017)

Also, more research is carried out by Hadjltaief using photocatalyst TiO_2/clay to degrade anionic reactive blue 19 (RB19) (Hadjltaief et al., 2019). The use of clay (loam) as a supporting material for TiO_2 leads to an increase in the specific surface area of the material from 36.6 to 116.7 m^2/g due to the distribution of TiO_2 particles. Characterization of TiO_2/clay using adsorption-desorption isotherms of N_2 is shown in Figure 9.

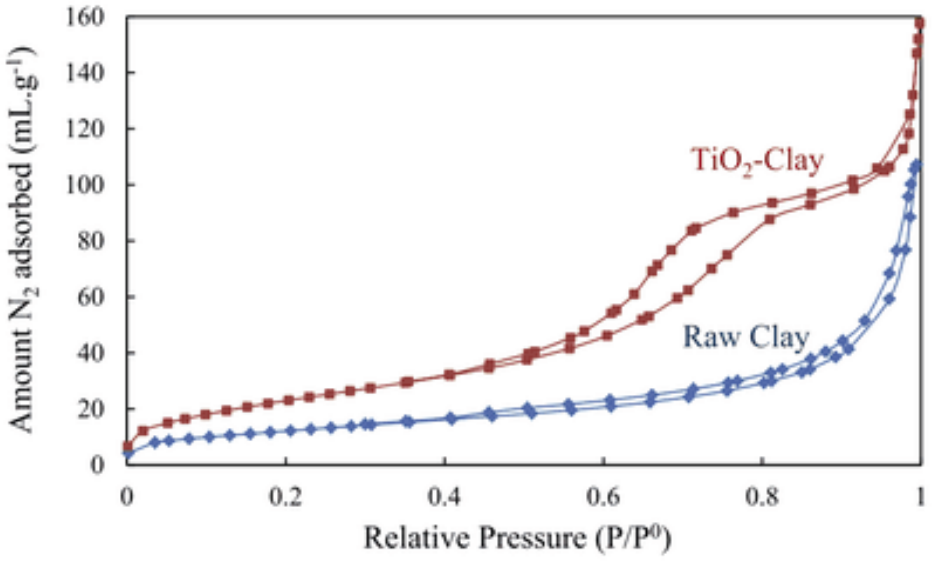

Figure 9. N_2 adsorption-desorption isotherms raw clay and photocatalytic TiO_2/clay (Hadjltaief et al., 2019)

6. Photocatalytic Activity of Humic Acid Degradation

Photocatalytic is a potential method used to degrade organic pollutants such as humic acid in peat water. This method has properties that are environmentally friendly, practical, and economical. The ability of the photocatalyst material in electron-hole recombination can transform organic pollutants into environment-friendly compounds and can be used repeatedly. The previous researchers have a lot to develop photocatalytic materials for use in degrading humic compounds, as shown in Table 5.

Table 5. Previous research on humic acid degradation

No.	Catalyst	Reaction Time (minutes)	Degradation Efficiency (%)	Reference
1.	Fe-TiO_2/activated carbon round	360	≤60	(Baek et al., 2013)
2.	TiO_2 thin layer	240	70	(Ramadhani et al., 2017)
3.	TiO_2 beads	240	80	(Andayani & Bagyo, 2011)
4.	TiO_2-Zeolite	300	80.41	(Julius, 2016)
5.	Ag/ZnO	40	88	(Ghaneian et al., 2014)
6.	TiO_2	300	89.4	(Jayadi et al., 2014)
7.	TiO_2/granular activated carbon	180	> 90	(Xue et al., 2011)
8.	TiO_2/activated carbon	60	≥95	(Kim et al., 2016)
9.	TiO_2	180	100	(Dziedzic et al., 2010)
10.	Cu-doped ZnO	120	100	(Maleki et al., 2015)

TiO_2 is a semiconductor material that is most widely used as a photocatalyst. Previous researchers have developed this material with certain modifications to achieve optimal degradation. As performed by Baek et al. about the photodegradation of humic acid using Fe-TiO_2/activated carbon with variations percent by weight (Baek et al., 2013). TiO_2 is modified by the addition of doping Fe and supporting materials such as activated carbon. This modification aims to enlarge the specific surface area of material that may improve photocatalytic performance. The isotherm adsorption and photocatalytic performance of this material are shown in Figures 10 and Figure 11.

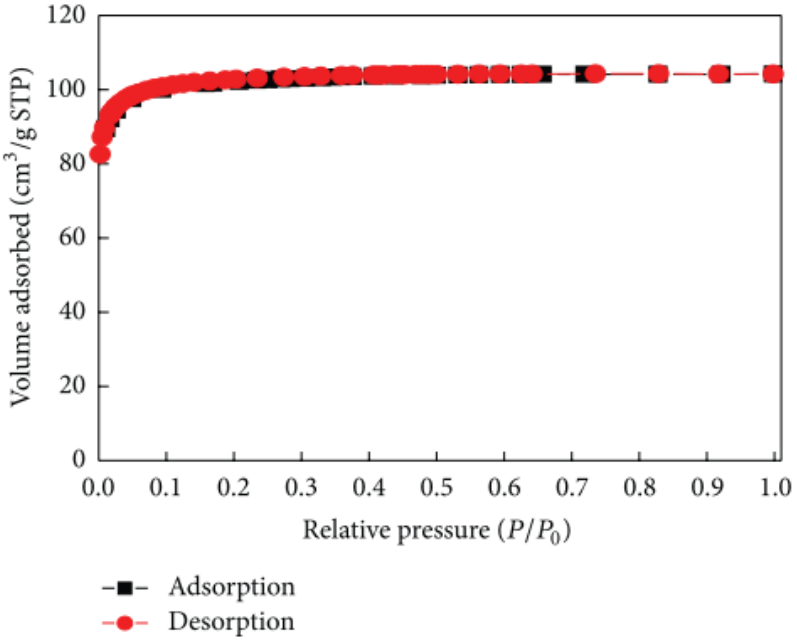

Figure 10. N_2 adsorption-desorption isotherms Fe-TiO_2/spherical activated carbon (0.6% wt) (Baek et al., 2013)

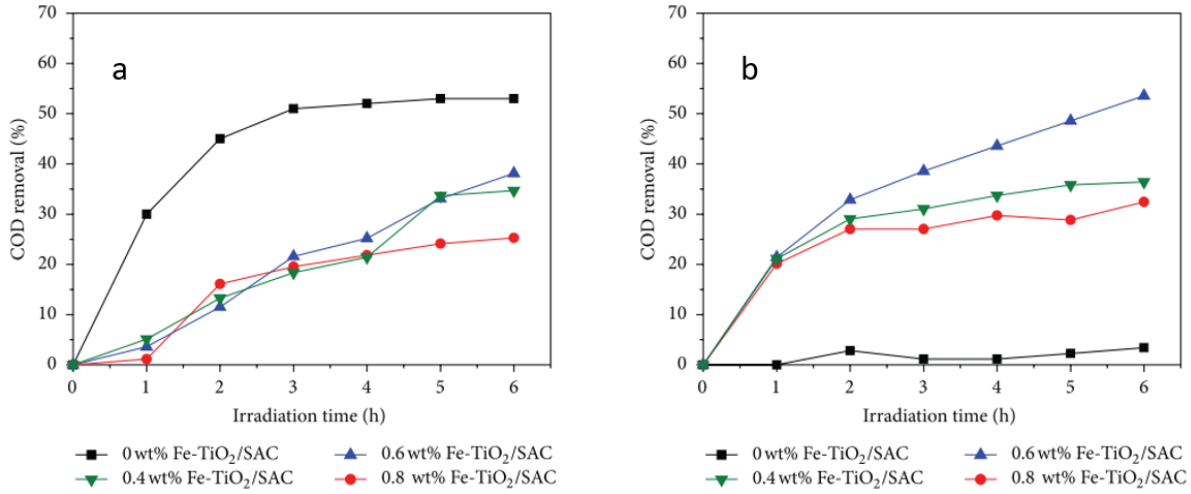

Figure 11. Results degradation of humic compounds under radiation with a wavelength of (a) 100~280 nm and (b) 315~400 nm (Baek et al., 2013)

The results of the characterization of Fe-TiO_2/spherical activated carbon (0.6% wt) using adsorption-desorption isotherms of N_2 are shown in Figure 10. The Fe-TiO_2/spherical activated carbon (0.6 wt%) has a surface area of 487 m^2/g. This material is used to degrade humic compounds under radiation with a wavelength of 100-280 nm and 315-400 nm, respectively for 6 hours. Based on Figure 11, it can be seen that the humic acid degradation under radiation with a wavelength of 100-280 nm is more optimal than the wavelength of 315-400 nm) (Baek et al., 2013).

Other studies on the degradation of the humic acid to achieve maximum results were conducted by Maleki et al. using a Cu-doped ZnO material (Maleki et al., 2015). Cu-doped ZnO synthesized by hydrothermal method. This material is used as a photocatalyst to degrade humic compounds with weight percent variation in the time interval of 120 minutes. The results of the highest degradation (100%) were obtained by Cu-doped ZnO material by weight percent of 1.5%.

7. Conclusion

Some photocatalyst has been developed by researchers all over the world both for waste treatment and other waters. The ability of the photocatalyst material in electron-hole recombination can transform organic pollutants into environment-friendly compounds. It can be used repeatedly to make photocatalytic is a method that could potentially be used for peat water treatment, especially to degrade humic compounds. There are many types of photocatalysts developed materials, either a metal oxide or perovskite, the combination of photocatalysts and adsorbents. Metal oxides are the most widely used as a photocatalyst is TiO_2 because of the nature of their thermal stability and its reusability. Also, materials such as ZnO, ZrO_2, ZnO_2, and others are often used as photocatalysts. The perovskite played a role as well as a photocatalyst, such as $LaFeO_3$, $LaNiO_3$, $BiFeO_3$, $BaTiO_3$, and much more. The development of the photocatalyst material is applied by researchers to degrade humic compounds such as TiO_2 and ZnO are supported by activated carbon, zeolites or modified with metal dopants such as Fe, Ce, and C. These materials are applied to various variables, either percent weight of the catalyst, a source of photons up to the time of irradiation. The success of researchers in degrading humic compounds using photocatalysts can be seen in the degradation efficiency, especially achieved by more than 50% degradation. Thus, it makes excellent photocatalytic potential as an optimal method for peat water treatment, especially humic acid degradation.

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