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Plastic Degrading Nanomaterials via Photocatalysis

Nisha Loura [a], Manvender Singh [a] and Vikas Dhull [a#*]

[a] Department of Biotechnology Engineering, University Institute of Engineering & Technology, Maharshi Dayanand University, Rohtak-124001, Haryana, India.

Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

The decomposition of plastic material is very slow and takes several years. There are lot of conventional methods for plastic degradation which require a lot of energy and break down into different types of fragments. There are various plasticizers and other compounds that make the plastic particles more durable. The approach to using nanomaterials for plastic degradation allows for a significant reduction in time. The effective application of the photocatalytic degradation approach with UV and solar light irradiation can significantly decompose plastics. Photocatalytic degradation takes place through the reactive oxygen species generation. The present review suggests a thorough comprehension of the photocatalytic breakdown via nanomaterials such as TiO_2, ZnO, and their nanocomposite as photocatalysts by determining the weight loss. These nanomaterials degrade various polymers like polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), low-density polyethylene (LDPE), and high-density polyethylene (HDPE) etc. In present work demonstrates nano-enabled strategies for microplastic and nanoplastic degradation.

[#] Assistant Professor;
**Corresponding author: Email: v.nano87@gmail.com;*

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1. INTRODUCTION

Plastic releases a variety of dangerous chemicals that damage aquatic life, pollute the environment, and lead to cancer and other serious ailments as it is a non-biodegradable material [1]. Because of its long durability and corrosion resistance, plastic trash lingers in the environment due to its difficulty in biodegradation and thus only breaks down into smaller bits [2]. Plasticizers and other chemicals enhance the durability of plastic particles [3]. Polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), low-density polyethylene (LDPE), and high-density polyethylene (HDPE), and polyethylene terephthalate (PET) are the synthetic organic polymers that make up the majority of plastics [4]. Bisphenol A, triclosan, organotin, brominated flame retardants, and phthalates are some of the plastic additives that pose risks to human health and the environment. Currently, landfilling, incineration, mechanical recycling, and chemical recycling are all utilized to manage plastic trash [5]. Mechanical processes basically breakdown the majority of plastics, and they might also cause the development of numerous microplastics and micropollutants. Some popular techniques for plastic degradation include oxidative photodegradation, thermal deterioration, biodegradation, catalytic decomposition, mechano-chemical degradation, and ozone-induced degradation [6].

Plastics naturally degrade very slowly and thus break down into different types of plastics, including mega plastics, macroplastics, mesoplastics, microplastics, and nano plastics with particle size diameters of >1 m, >2.5cm, 0.5-2.5cm, 1 µm–5 mm, and <1000 nm, respectively [7-9]. In marine, freshwater, oceanic, and terrestrial ecosystems, nano plastics and microplastics are extensively dispersed [10].

Thermal, conventional, and chemical processes require a significant amount of energy for plastic degradation, and they also leave behind a sizable number of issues once the degradation process is complete. By changing the microbes metabolic cycles, nanoparticles (NPs) can improve their ability to degrade polythene [11]. Materials that have at least one exterior dimension that is between 1 and 100 nm in size are considered nanomaterials [12]. With their exceptional characteristics, such as a large surface area and increased porosity, nanomaterials promote plastic degradation while having outstanding physicochemical stability [13]. Weight loss can be used to gauge the deterioration of plastics by using Fourier-transform infrared spectroscopy (FTIR) and Scanning electron microscopy (SEM) to confirm morphological and chemical changes [14].

2. MECHANISM OF PHOTOCATALYTIC PLASTIC DEGRADATION

An advanced oxidation process (AOP) called photocatalytic degradation employs light energy to hydrolyze pollutants when photons are absorbed [15]. The photocatalytic process takes place during the electrons transition from the valence band (VB) to the conduction band (CB) while the semiconductor is bombarded with photons that generate a hole (h^+) in the VB. The radicals of superoxide ($O_2^{\bullet-}$) and hydroxyl (·OH) are created by the interaction of these photogenerated moieties with water and oxygen adsorbed on the semiconductor surface. The (·OH) radical contributes to the breakdown of plastics [16]. These reactive oxygen species (ROS) are potent oxidising agents that have the potential to convert organic contaminants that have been deposited on the semiconductor surface into CO_2 and H_2O [17]. ROS are produced by semiconducting materials and operate as catalysts in photocatalytic processes, which promise to be economical and environmentally benign ways to degrade plastics [18]. The most often used photocatalyst components are nano-semiconductors. To be an effective photocatalyst, semiconductors must have a number of desirable characteristics, including chemical and biological stability, lack of toxicity, affordability, strong UV-visible activity, and photo responsiveness [19]. The catalyst type, temperature, light duration and intensity, pH, etc. all have an impact on the photocatalytic degradation of nanoplastics and microplastics [20]. When plastics degrade, there are two basic mechanisms involved: direct and indirect degradation [9]. The mechanism for degradation of plastics is shown in Fig. 1.

3. PHOTOCATALYTIC PLASTIC DEGRADATION VIA NANOPARTICLES

Photocatalytic degradation process effectiveness will be constrained if the irradiation of light is blocked by objects, as it is a surface phenomenon [21]. As a result of their widespread

availability, low toxicity, and low cost, zinc oxide (ZnO) and titanium dioxide (TiO_2) nanoparticles (NPs), in addition to their complexes, are some of the most frequently usable photocatalysts [22]. TiO_2 has a energy band gap of 3.2 eV, which restricts the amount of Ultraviolet energy it can absorb [23]. In the UV region of the spectrum, the photon energy of wavelength (λ) around 388nm is required for TiO_2 photoexcitation. As a result of TiO_2 deactivation, abundant attempts have been done to further enhance its photocatalytical abilities. These include adding impurities to TiO_2, designing defects into the material's surface and bulk, synthesizing high energy facets of TiO_2 NPs, and creating composite materials using TiO_2 and graphene oxide (GO). These initiatives seek to increase the effectiveness of adsorption of TiO_2 and separation of e^- and h^+ [24]. TiO_2-based nanomaterials exposed to solar radiation can degrade polypropylene films. To compare the photodegradation of polypropylene earlier and later after 130 hours of solar irradiation, PP has been mixed with TiO_2 nanoparticles and its nanocomposites i.e., TiO_2-reduced graphene oxide (TiO_2-rGO). An effective enhancement of TiO_2-rGO nanocomposite based photodegradation of polypropylene has been revealed by microstructural and structural analysis, as determined by the emergence of cavity of 500nm diameter by FTIR and calculations of the carbonyl index by FE-SEM spectroscopy. Time of Flight-Secondary Ion Mass Spectrometry chemical state analysis demonstrates the formation of reactive oxygen species and subsequent photodegradation [25]. The examples of various nanomaterials in the photocatalytic degradation of platicizers are given in Table 1.

However, under UV and artificial solar radiation, aluminium-doped TiO_2 nanoparticles have demonstrated improved photocatalytical deterioration of a combination of plasticizers, i.e., 4-tert-butylphenol (4tBP) and bisphenol A (BPA). The sol-gel technique with microwave assistance has been used to create an Al-doped TiO_2 photocatalyst. At 4.9 pH, the Al-doped TiO_2 photocatalyst (0.05%) has shown improved photocatalytic activity, enabling a 61% and 55% rate of degradation for 4tBP and BPA in 3 hours under ultraviolet illumination, respectively [26]. Besides, Titania has been used as a photocatalyst to facilitate the visible light-induced photocatalytical polyethylene degradation. Titania nanotube sensitization with 10% brilliant green dye on polyethylene films has shown degradation of about 50% in 45 days under visible light. Film flaws caused by the formation of volatile organic products during photodegradation are validated by measuring tensile strength, FTIR, and SEM [27]. Titania (TiO_2) NPs of 50 nm have also degraded polyethylene film by an 18% breakdown rate under UV light [28]. By using a nanocomposite photocatalyst of polypyrrole/TiO_2 (PPy/TiO_2) through emulsion polymerization and sol-gel methods, polyethylene plastic has been photocatalytically degraded under sunlight radiation. It has been shown that exposing the PE plastic to sunlight for 240 hours results in weight reductions of pure TiO_2 and PPy in comparison to PPy/TiO_2 nanocomposite by 35.4% and 54.4% of Mw, respectively [29]. The study conducted by Zhao et al. has shown PE plastic degradation by solid-phase photocatalytically with TiO_2 NPs in the open air under the influence of solar and UV rays. 42% weight loss after 300 hours of solar irradiation has been observed in PE-TiO_2 film. By adjusting the TiO_2 NPs amount in the composite films of PE-TiO_2, the rate of their decomposition may be managed [30]. In another study, sunlight-active TiO_2 adorned magnesium doped cadmium ferrite nanocomposite (CdMgFe2O4@TiO_2) has shown effective removal of bisphenol A and tetrabromobisphenol A (TBBPA) plastic additives. At neutral pH and in the presence of sunlight, CdMgFe2O4@TiO2 has demonstrated maximal degradation of about 91% for TBBPA and 94% for BPA. The observations have been corroborated by a negative zeta potential, photoluminescence, a narrower band gap, and a high surface area. Numerous hydroxyl radicals target less protected sites and quickly destroy the additives in plastic [31].

Although TiO_2 has received greatest interest as a semiconductor nanomaterial for the organic molecules photocatalytical decomposition, ZnO has recently drawn greater awareness as TiO_2 potential substitute. In comparison to TiO_2, ZnO has a larger absorption capability and a wider UV spectrum of solar light despite having the same energy band gap [32]. The band gap of ZnO is 3.2 eV as similar to TiO_2. ZnO also absorbs a greater proportion of the solar spectrum and UV light wavelengths [33]. ZnO nanoparticles have the potential of photocatalytic degradation attributable to their excellent ultraviolet absorption transparency and efficacy [34]. Kamalian et al. have demonstrated the photocatalytic degradation of LDPE film by ZnO NPs grafted with two quantities of polyacrylamide

by describing the shape, mechanical characteristics, and weight reduction of the films under UV radiation. After 200 hours of exposure, the amount of ZnO NPs in LDPE sheets accelerated deterioration. The results of experiments utilising thermogravimetric analysis and FTIR provide additional proof that grafting polyacrylamide on the ZnO NPs in amounts of 10 and 39 weight percent has been successful. After 200 hours of exposure, the amount of ZnO nanoparticles in LDPE sheets accelerated deterioration [35].

Under sunlight exposure, ZnO NPs cause LDPE film to degrade. The photocatalytic degrading property of ZnO NPs has improved the degradation rate and overall weight loss by 67% after irradiation duration of 300 h. After 45 days, ZnO NPs treated LDPE films exposed to sun radiation had their greatest loss of tensile strength and elongation [36]. In another study, iron-doped ZnO (Fe-ZnO) NPs synthesis by the co-precipitation method has shown the visible-light-induced photocatalytic destruction of pure LDPE and commercial-grade PE films. Since no deterioration has been seen in the dark, the deterioration of 6.35cm^2 of commercial-grade PE and LDPE films is light dependent by producing ROS from the surface of nanoparticle. The results revealed commercial grade PE has a maximum reduction in weight of 15%, and pure LDPE films have a reduction of 13.8% in 14 days [37].

4. NANO-ENABLED APPROACHES FOR MICROPLASTIC DEGRADATION

Primary and secondary microplastics are the two categories of microplastics [38]. Polystyrene, polyethylene, and polypropylene particles are the primary sources of microplastics in pharmaceutical and cosmetic products. Secondary microplastics are created when plastic trash is broken up by biological, physical, and chemical processes [39]. Degradation primarily modifies the chemical and physical characteristics of primary and secondary microplastics, such as density, surface morphology, crystallinity, colour, and particle size [40]. The techniques that are most frequently used for locating and measuring microplastics include FTIR [41] and Raman spectroscopy [42].

Researchers Ariza-Tarazona et al. have used the temperature and pH roles in HDPE degradation in microplastics under visible light have been achieved using carbon and nitrogen-doped TiO_2 (C,N-TiO_2) nanoparticles. Low pH introduces H^+ ions that promote increased contact between the microplastics containing HDPE and colloidal NPs, which speeds up the decomposition of plastic. Microplastics become fragmented at low temperatures, which increases their surface area and enhances their C,N-TiO_2 interaction. The typical mass loss observed after 50 hours at pH 3 and 0°C temperature is 71.77 ± 1.88% [43]. Haris et al. have synthesized silver-doped TiO_2 photocatalyst that has shown improved performance in destroying microplastics in bottle-sourced water samples via photoassisted deposition approach. The results demonstrate that the catalyst may degrade microplastic compounds in water when exposed to UV light, with a 3% Ag/TiO2 catalyst demonstrating the most potent degradation capability after 4 hours of exposure. After deterioration, only 9.5 mg of microplastics left, which represents an 81% mass degradation percentage [44].

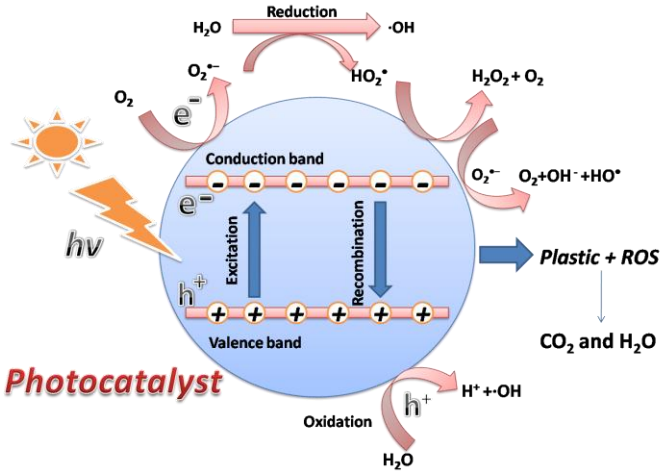

Fig. 1. Diagrammatic representation of plastic degradation mechanism via photocatalysis

Table 1. Nanomaterials in plastic degradation via photocatalysts

Nanomaterials	Plastisizer	Synthesis Method	Light Source	Catalysis time	Weight reduction	Reference
Al-TiO_2	Bisphenol A (BPA) and 4-tert-butylphenol (4tBP)	Microwave-assisted sol-gel method	UV light (λ = 365 nm)	3 h	55% (4tBP) and 61% (BPA)	[26]
TiO_2 coated magnesium doped cadmium ferrite ($CdMgFe_2O_4$@TiO_2	tetrabromobisphenol A (TBBPA) and bisphenol A (BPA)	*M.koenigii* plant extract via co-precipitation	Sunlight	TBBPA ($t_{1/2}$:2.4 h) and BPA ($t_{1/2}$:2.1 h)	TBBPA (91%) and BPA (94%)	[31]
ZnO NPs	LDPE films	Green synthesis areca nut (*Areca catechu*) extract and	Solar irradiation	300h	67%	[36]
TiO_2 NPs	Polystyrene (PS) microspheres	N/A	254 nm nm UV Light	24 h	complete mineralization (98.40%)	[45]
ZnO nanorods	Polypropylene microplastics	N/A	Visible light	456 h	65%	[48]

Note: "N/A" represents not available

Besides, some research has suggested that microplastics like polyethylene (PE) film and polystyrene (PS) microspheres might be effectively degraded and completely mineralized by a coating of TiO_2 nanoparticles when exposed to UV light. Using Triton X-100 and TiO_2 nanoparticles, it has shown that 98.40% of the 400 nm PS completely mineralized in 12 hours. After 36 hours, the experiment for PE degradation also showed a strong rate of photodegradation. The degradation method revealed by in situ mass spectrometry and diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) studies has provided the formation of carbon-hydrogen, hydroxyl, and carbonyl groups during PS photodegradation [45]. Allé et al. have shown the ability to mineralize polymethylmethacrylate and polystyrene nanobeads using TiO_2 in an aqueous solution under UV light. For the removal of polymethylmethacrylate microplastics, TiO_2-P25/-SiC foams have been used in a flow-through system and achieved a 50% TOC conversion after 7 hours of treatment at an irradiation of 112 W/m^2. According to the study, the photocatalytic method works well with polymers with a variety of molecular architectures, including polystyrene and polymethyl methacrylate, as well as with a range of typical nanobead sizes [46].

Apart from TiO_2 nanoparticles, zinc oxide nanorods have also been synthesized for microplastic residues and low-density polyethylene (LDPE) degradation via photocatalysis under visible light. The residues carbonyl index observed to be increased by 30%, resulting in the appearance of more cavities, creases, and cracks on LDPE surface along with increased brittleness [47]. However, Uheida et al. have synthesized ZnO nanorods for polypropylene microplastic residue degradation under visible light by a photocatalytic mechanism. After 456 hours of exposure to visible light, the FTIR data show that PP microplastics have been photodegraded effectively due to the emergence of carbonyl groups with a higher carbonyl index led to a 65% decline in the average particle volume. Flaws in PP are caused by the formation of volatile organic products during photodegradation, and these flaws are supported by SEM and FTIR tests [48]. The study conducted by Tofa, Ye, et al. uses plasmonic photocatalysts induced by visible light made of platinum NPs coated on ZnO nanorods (ZnO-Pt) to show the low-density polyethylene film degradation in microplastics. As a result of plasmonic catalyst synergistic effects, the photocatalytic activity under irradiation with visible light is increased by more than 15%. ZnO-Pt catalysts, in comparison to ZnO nanorods, have demonstrated an elevated potential for oxidising LDPE film. The cause of LDPE films photocatalytic degradation is ensured by oxygenated group formation such as peroxides, unsaturated groups, and hydro-peroxides that cause holes, wrinkles, and breakage [49].

5. NANO-ENABLED APPROACHES FOR NANOPLASTIC DEGRADATION

Due to their distinct physiochemical nanoscale features, nanoplastics may be more hazardous and pose greater dangers when compared to microplastics [50]. The nanoplastics production and degradation processes can be studied using nanoparticle tracking analysis [51]. There are two classifications of sources for nanoplastics include: primary sources, such as nanoscale plastics as a result of improper product disposal entering the environment, and secondary sources via the decomposition of macroplastics and microplastics [52]. Microplastics found in soil and water degrade and fragment, producing nanoplastics as a byproduct [53]. As a result of their large surface area and small size, nanoplastics are a hazard for the environment as they encourage the adsorption and transit of other pollutants, including heavy metals and polycyclic aromatic hydrocarbons, via the food chain [54]. Understanding the cost of nanoplastic availability and its biological consequences requires an understanding of biological reactivity, which enhances when plastic particle size reduces [55]. Generally, nanomaterials are frequently created on purpose because of their distinctive features, but they can also accidentally end up in various manufactured goods, like powders, because of the particle size distribution. Nanoplastics are a particular class of nanomaterials that are typically found as byproducts of the deterioration of plastic polymers [56].

Domínguez-Jaimes et al. have synthesized three different types of photocatalysts based on TiO_2, i.e., nanotubular (TiO_2/T), mixed (TiO_2/M), and barrier (TiO_2/B), via the electrochemical anodization method for degradation of polystyrene nanoplastics with UV light. The findings demonstrated that TiO_2/M, with its multilayer structure containing nanotubular and nanograss morphologies, outperformed the other synthetic photocatalysts. The TiO_2/M structure

and nanoplastic interface have led to a 23.5% drop in pollutant concentration [16].

6. RATIONAL DESIGN AND FABRICATION OF HIGH EFFICIENT PHOTOCATALYSTS

Due to their electrochemical stability, affordability, and redox capability, metal oxide semiconductors, such as TiO_2, are intensively explored. Three processes—photodeposition, hydrogenation, and photoreduction—are required to create the nanoheterostructure. X-ray diffraction analysis can be used for the identification of photocatalyst crystal structures. To describe the chemical compositions of surfaces, X-ray photoelectron spectra and transmission electron microscopes for the characterization of the elemental and microstructure arrangements can be used. To examine the photocatalysts' light responses, UV-vis absorbance spectroscopy can be used. To learn more about the structural alterations made during the creation of the nanoheterostructures, Raman spectra can be measured. These methods may offer fresh perspectives on designing metal oxide semiconductors with a wide bandgap and improved transport and separation spatial charge as well as photocatalytic light absorption capabilities [57].

Hydrothermal fabrication has been used to create nanocomposite TiO_2-ZnO photocatalysts [58]. According to reports, TiO_2 can expand its photoresponse into the visible light area by forming a heterojunction with other semiconductors. ZnO modification of TiO_2 is thought to be a potential method that might enhance charge transfer and decrease recombination of photogenerated electron-hole pairs that possess the ability to photodegrade [59]. Apart from hydrothermal fabrication methods, other methods include sol-gel, solvothermal, calcinations, immobilization, chemical precipitation, electrochemical anodization, template-based, chemical vapor deposition, sonochemical and microwave-assisted methods [23,60]. A nanocomposite of cellulose acetate CA@TiO_2 has been created by electrospinning a polymeric matrix to create a TiO_2 NP core, which has then been submerged in a suspension solution of ZnO NPs for deposition of ZnO NPs on the surface of the nanofiber. The composite mat has then been put through a calcination/annealing procedure at 700°C for 4 hours to produce nanocomposite heterojunction nanostructure material. Due to the highest ceramic content, the produced hetrojunction TiO_2@ZnO nanoparticles have strong thermal characteristics [61]. In another study, the production of ZnO core-shell nanofibers wrapped in graphene oxide (GO) (ZnO@G CSNFs) has been synthesized using a straightforward core-shell electrospinning process at a rate of 0.01 mL/min and subsequent annealing/calcining in air at 450°C for 3 h for an effective photocatalytic process and enhanced stability. The GO has been placed on the surface of the heterostructured catalyst, acting as a protective shell, with ZnO creating an enclosed core component [62].

7. INDUSTRIAL APPLICATION OF PHOTOCATALYSTS

The principal application of photocatalytic technology nowadays is to clean industrial effluent, such as that produced by the paper, printing, dyeing, and electroplating industries [63]. A significant procedure must be used to remove the undesirable contaminants because the buildup of organic compounds and dyes is hazardous to the aquatic species that already present. The most current method uses TiO_2 in particular as a photocatalyst for photocatalysis using several kinds of metallic-oxide photocatalysts [64]. The photocatalysis for disinfection shows that this method has the potential to be widely used in a variety of fields, including drinking water disinfection, medical, biological, pharmaceutical, hospital applications, laboratory, and food industry applications [65]. Nanomaterials including ZnO, TiO_2, and nanoclay have made it possible to sequester a range of contaminants from wastewater. TiO_2 and ZnO anchored on clay have been investigated for wastewater cleanup using nanotechnology [66].

8. CHALLENGES AND FUTURE PERSPECTIVES

The photocatalytic breakdown of plastics (microplastics and nanoplastics) has several advantages, but there are still certain issues that need to be resolved. The specificity of photocatalysts, or the fact that they are frequently suitable for degrading only a specific type of plastic, is one of the significant restrictions. Additionally, photocatalysts are unable to specifically target the reactive sites in plastic [21]. An obstacle could be the degradation of plastics and the co-pollutants utilising photocatalytic degradation. It has been

determined that the microplastics are coupled with other environmental co-pollutants; appropriate methods for reducing combined pollution should be taken into account. This review has covered recent developments in TiO_2 and ZnO NPs based photocatalysts as well as the creation of fabrication methods for plastic degradation. However, the fundamental issue with TiO_2 and ZnO is their large bandgap, which leads to photo-induced excitations to quench quickly. This lessens TiO_2 and ZnO capacities for photocatalysis. Additionally, it is necessary to research how the synthetic processes affect photocatalytic activity. Even though the described TiO_2 and ZnO nanomaterials have demonstrated great photocatalytic activity, more experimental and theoretical research is needed to fully comprehend and advance the development process of plastic degradation. Future research and development on the photocatalytic degradation of plastics can concentrate on controlling the morphology of nanomaterial-based photocatalysts with high stability and the creation of other unique nanomaterial-based photocatalysts to degrade microplastics and especially nanoplastics, as the available information is insufficient.

9. CONCLUSION

Due to ineffective recycling, the degradation of non-biodegradable plastics has drastically increased**.** The plastic granules are strengthened by a variety of plasticizers and other substances. Plastics can be significantly decomposed when the photocatalytic degradation method is effectively used under irradiation with UV and solar light. This review makes a strong case for a complete understanding of the photocatalytic breakdown of plastic utilising nanomaterials like TiO_2, ZnO, and their nanocomposite as photocatalysts by measuring the weight loss. Numerous polymers, including high-density polyethylene, low-density polyethylene, polypropylene, polystyrene, and polyvinyl chloride, among others, are degraded by these nanomaterials. Microplastics and nanoplastics come from both primary and secondary sources. The present review has demonstrated the photocatalysis mechanism along with the role of TiO_2 and ZnO nanomaterials in the degradation of various polymers and plasticizers associated with microplastics and nanoplastics. However, there is still a requirement for other nanomaterials for these plastic degradations as the current studies are still lacking.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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