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## Recycling waste aluminium foil to bio-acceptable nano photocatalysts [aluminium oxide ( $\text{Al}_2\text{O}_3$ ) & aluminium oxyhydroxide ( $\text{AlOOH}$ )]; dye degradation as proof-of-concept†

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The surge in the world's population resulting from urbanization and industrialization has led to a significant uptick in water and soil pollution. Aligned with the United Nations' sustainable development goals, investigating innovative methods for repurposing waste into beneficial materials and effective catalysts that are compatible with ecosystems and capable of efficiently decomposing dyes is earnestly recommended. Additionally, in alignment with the objectives of a sustainable society, this study serves as a prototype for repurposing discarded aluminium foil—an everyday single-use material contributing to landfill accumulation—into aluminium oxide ( $\text{Al}_2\text{O}_3$ ) and aluminium oxyhydroxide ( $\text{AlOOH}$ ) nanocatalysts, intended for efficient photodegradation applications.  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  nanosystems were synthesized using a well-optimized chemistry route. The developed nanosystems were characterized using FTIR, EDX mapping, XRD, FE-SEM, and TGA/DTA that found the bonds, composition, structure, morphology of the particles, and thermal stability, respectively. These particles were used for the degradation of cationic methylene blue (MB) dye in neutral (pH 7), basic (pH 9), and acidic (pH 5) mediums. Liquid chromatography mass spectrometry (LC-MS) was performed to check the MB intermediate product formation on photodegradation. The findings suggest that exposing cationic MB to light in neutral pH conditions with  $\text{Al}_2\text{O}_3$  is highly effective, with a dye degradation rate of 99.29%. Exposing MB to the dark in neutral conditions with  $\text{AlOOH}$  is the least effective, with a dye degradation rate of 6.64%. As the pH is made more acidic and/or basic, the effectiveness of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  also slightly changes. The outcomes related to reusability and toxicity studies also proved the acceptability of the developed systems. Degradation using both compounds led to more germination when compared to MB, and both compounds showed outstanding reusability. The research emphasizes the importance of sustainable materials synthesis and offers valuable insights for the development of efficient photocatalysts tailored for specific environmental conditions in the context of dye degradation.

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## 1. Introduction

Over the past century, the global population has surged from 2.2 billion individuals to 7.9 billion, showing an exponential growth trend due to technological advancement and industrialization. Consequently, this expansion has led to the generation of new pollution sources, including solid waste pollution in water. The interconnection between population growth and pollution is clear in the widespread increase in single-use products and other disposable packaging materials. The global pandemic has further propelled industrial demand for single-use materials. Since the COVID-19 pandemic (2020), single-use food packaging containers and wrappers have drastically increased.<sup>1,2</sup> As the demand for these products rises, environmental degradation caused by the production of raw materials

and the disposal of old materials also escalates. One such waste-generating, single-use pollutant is aluminium foil, whose development is intricately linked with industrialization and a rise in food production. Aluminium foil is an integral part of the packaging industry, including food preparation and several other industrial applications; nevertheless, aluminium disposal poses a significant threat to the environment.<sup>3</sup> Since the lockdown in 2020, there has been a 54% increase in food wrapper use globally.<sup>1,2</sup> Aluminium pollution stems from the widespread improper disposal of aluminium-based materials and is linked to natural and anthropogenic sources. If aluminium is disposed of incorrectly, it creates environmental degradation regardless of whether it decomposes or not. If the foil decomposes or breaks down, it will introduce new chemical compounds to the environment, causing environmental pollution. However, if the foil does not break down, it further causes solid waste pollution in waterways and other ecosystems. These anthropogenic sources have a considerable impact on environmental and human health.

When aluminium foil is correctly disposed of, it ends up in landfills, incinerators, or recycling plants. However, the foil has a long decomposition cycle, causing it to persist in municipal solid waste facilities for extended periods.<sup>4</sup> The incineration of foil causes increased air pollution and further causes soil and water pollution.<sup>5</sup> Aluminium in landfills poses equally threatening environmental problems by generating unwanted heat from chemical reactions during decomposition, producing liquid leachate, and releasing gases such as hydrogen, hydrogen sulfide, carbon monoxide, and ammonia, all of which have considerable impacts on human health.<sup>5,6</sup> Trends indicate an increase in diseases such as Alzheimer's disease, autism, epilepsy, and other neurological diseases associated with these pollutants.<sup>7</sup> Aluminium is also difficult to recycle, as the foil that is finally brought to recycling facilities is generally torn and wrinkled or contaminated with food debris. Compromised foil cannot be recycled. Solid aluminium waste is a concern for municipal solid waste facilities and environmental health and safety management due to its persistent nature. However, if waste foil is treated to remove contaminants, then it can be used to synthesize aluminium particles that can be used multifariously in dye degradation, absorption, dye binding, as desiccating agents, in drug delivery, for antimicrobial potential, and as photocatalysts in waterways.<sup>8-10</sup> It has been reported that various metal oxides and nanocomposites have been prepared and used for sensing, energy storage and the removal of pollutants in water.<sup>11-13</sup> Studies have shown promising results for  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  particles in the degradation of organic pollutants for the treatment of contaminated water.<sup>14</sup> Specifically, many photodegradation reactions have been performed using  $\text{Al}_2\text{O}_3$  as a photocatalyst. A recently published review article meticulously scrutinises recent developments in  $\text{Al}_2\text{O}_3$ -based materials, highlighting their efficacy in organic dye adsorption and degradation.<sup>15</sup> For example, Anna *et al.* used these particles as a photocatalyst for the degradation of methylene blue (MB) under sunlight.<sup>16</sup> In another study, the photocatalytic properties of  $\text{Al}_2\text{O}_3$  were explored to degrade ciprofloxacin in

wastewater.<sup>17</sup> Zang *et al.* investigated  $\text{AlOOH}$  as a photocatalyst for the degradation of tetracycline hydrochloride.<sup>18</sup>

These aluminium particles are significant in building a sustainable future as population growth and pollution continue to threaten global water supplies. Industrialization further causes a massive influx of dyes and other synthetic chemicals into our waterways; globally, there are about 60 000 tons of waste dye that is released into the environment per year.<sup>19,20</sup> The textile industry is largely responsible for this pollution; however, the removal of dye from wastewater is an incredibly strenuous and expensive process. Methylene blue is a vibrantly colored, cationic dye that is commonly used in many industries, such as textiles, pharmaceuticals, and microbiology (for cell staining).<sup>11,16</sup> The versatility of this dye lies in its ability to solubilize in a variety of solvents, such as water, ethanol, and acetic acid.<sup>16,21</sup> This dye is very harmful to marine life as well as humans in higher concentrations.<sup>21</sup> Anaerobic reduction, microfiltration, membrane filtration, and other techniques have been employed to try to degrade MB dye.<sup>22-24</sup> So far, it has been proved that adsorption is an effective technique for removing the dye from polluted water.<sup>22</sup>

Based on the approach of recycling waste to wealth, this research for the first time focuses on the remediation of water containing MB dye by degrading it using photocatalytic particles that were prepared from waste aluminium foil. Synthesized aluminium oxide ( $\text{Al}_2\text{O}_3$ ) and aluminium oxyhydroxide ( $\text{AlOOH}$ ) nanocatalysts were fabricated and characterized using Fourier transform infrared spectroscopy (FTIR), energy-dispersive X-ray spectroscopy (EDX) mapping, X-ray diffraction (XRD), field emission scanning electron microscopy (FE-SEM), UV-vis diffuse reflectance spectroscopy (UV-DRS), and thermogravimetric analysis/differential scanning calorimetry (TGA/DSC).

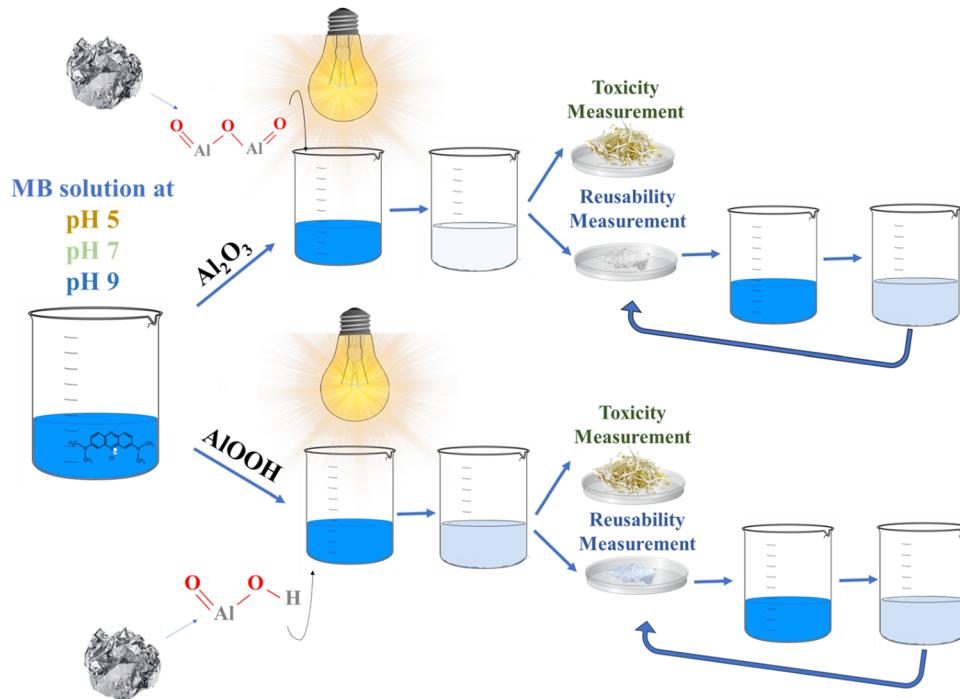
These particles were further used for the degradation of cationic MB in neutral, basic, and acidic pH mediums. The particles were checked for reusability and toxicity assessment. This research addresses multiple goals simultaneously: a reduction in foil solid waste and the production of aluminium particles for water pollution removal. Additionally, because the dye is decomposed *via* visible light, sunlight can be used for the pigment degradation; thus, no additional energy source is required. Scheme 1 shows a graphical representation of this work.

## 2. Experimental details and procedure

### 2.1. Materials

Aluminium foil was collected from lab waste sites at Panjab University, Chandigarh. Hydrochloric acid (HCl) and sodium hydroxide (NaOH) pellets were procured from ThermoFisher; sodium carbonate anhydrous LR ( $\text{Na}_2\text{CO}_3$ ) was bought from Rasayan Laboratories. All chemicals were used in their original state without any later modification or purification. All experiments were conducted at room temperature in double-distilled water (ddH<sub>2</sub>O). A methylene blue (MB) dye was procured from Sigma-Aldrich. For toxicity assessment, locally sourced (Chandigarh) mung seeds were used.





Scheme 1 Graphical representation of the work.

## 2.2. Experimental procedure

**2.2.1. Synthesis of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$ .** Waste foil was collected and put in a hot-water bath for one hour to remove any earlier contamination. 3.89 g of foil was then shredded into small, approximately 2–3 cm pieces and added into a 1:1 solution of 23 mL of HCl and 23 mL of ddH<sub>2</sub>O. This forms an aluminium chloride ( $\text{AlCl}_3$ ) solution. Once all the foil was completely dissolved into the HCl, the  $\text{AlCl}_3$  solution was double filtered into a separate beaker to remove any remaining impurities. This solution was divided into two parts; each half was used to synthesize  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$ , respectively.

$\text{Al}_2\text{O}_3$  was synthesized by adding excess  $\text{Na}_2\text{CO}_3$  into one of the two halves of the filtered solution until a gelatinous product was formed. This product was transferred into a large beaker, washed with excess ddH<sub>2</sub>O, decanted, and washed again. The decantation and washing were repeated twice. The remaining

water was disposed of after the second decantation, and the solid precipitate was dried in an oven at 45 °C for 24 hours and stored in a desiccator to remove any moisture content.

$\text{AlOOH}$  was synthesized by adding excess NaOH into the second half of the filtered solution until a gelatinous product formed. This product was transferred into a large beaker, washed with excess ddH<sub>2</sub>O, decanted, and washed again. The decantation and washing were repeated twice. The remaining water was disposed of after the second decantation, and the solid precipitate was dried in an oven at 45 °C for 24 hours.<sup>25</sup> Fig. 1 shows the overall step-by-step synthesis procedure from waste aluminium foil.

**2.2.2. Preparation of MB dye.** A 5 ppm MB dye was prepared as a bulk stock solution. The effectiveness of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  was evaluated under dark and light conditions in a neutral pH of 7, basic pH of 9, and acidic pH of 5. Basic pH conditions were created by the addition of NaOH pellets to a

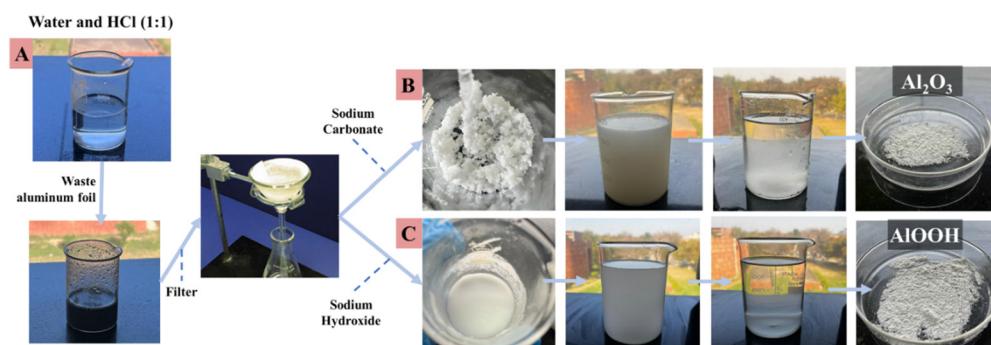


Fig. 1 Photographs of the synthesis procedure of  $\text{AlCl}_3$  (A),  $\text{Al}_2\text{O}_3$  (B) and  $\text{AlOOH}$  (C).

part of the bulk solution, while acidic pH conditions were created by adding HCl to another part of the bulk solution. The pH of the solution after the addition of NaOH and HCl was measured using a pH meter (Labman Scientific Instruments).

**2.2.3. Characterization.** Several instruments were employed to analyze the synthesized  $\text{Al}_2\text{O}_3$  and ALOOH particles. Chemical bonds in the samples were characterized using Perkin Elmer Spectrum II FTIR with a scanning range of  $4000\text{ cm}^{-1}$  to  $400\text{ cm}^{-1}$ . The morphology, topology, and metallographic details of the particles were observed using a Field Emission-Scanning Electron Microscope (FE-SEM) from the HITACHI SU8010 series. Solid material was cast on carbon tape and coated with a gold sputter coating machine at  $15\text{ mA}$  for 20 seconds. For mapping and EDX analysis, the operating voltage was  $15\text{ kV}$  and the working distance was  $15\text{ mm}$ . Elemental analysis was performed using an EDS-Bruker SDD XFlash 6130. An Anton Paar Litesizer 500 Zeta-sizer was used to measure the particle size and zeta potential (mV). The DLS experiments were performed on the instrument equipped with a front scattering angle of  $15^\circ$ , a side scattering angle of  $90^\circ$ , a back scattering angle of  $173^\circ$ , and a He-Ne laser (wavelength =  $633\text{ nm}$ , power =  $4\text{ mW}$ ). The thermal properties of the prepared particles were investigated using thermogravimetric analysis (TGA-DTA) on an SDT Q-600 instrument. The crystallographic structure was explored using an X'Pert pro X-ray diffraction (XRD) spectrophotometer. UV-vis diffuse reflectance spectroscopy (UV-DRS) (Jasco V-750) was used to calculate the band gaps of  $\text{Al}_2\text{O}_3$  and ALOOH.

**2.2.4. Photocatalytic activity experiments.** A total of  $25\text{ mL}$  of the dye stock at  $5\text{ ppm}$  was used along with  $50\text{ mg}$  of each particle for all experiments. The solutions with the photocatalysts were placed under dark and visible light conditions at pH 7, pH 9, and pH 5, for two trials each. A visible light bulb ( $500\text{ W}$  tungsten lamp) was used for this experiment and was placed approximately 12 inches above the beaker (containing the MB-contaminated water and photocatalyst particles). The absorbance of each solution was recorded at 30-minute intervals for 240 minutes using a UV-vis spectrophotometer (LABINDIA UV 3200). A quartz cuvette was used for the measurement. The following equation was used to calculate the degradation efficiency (DE) of the particles in the dye:

$$\text{DE} (\%) = \frac{C_t}{C_0} (100) \quad (1)$$

where DE is degradation efficiency as a percentage,  $C_0$  is the initial concentration and  $C_t$  is the final concentration of dye after time 't' of photodegradation.

The formation of methylene blue intermediates after photocatalytic degradation was analysed using a mass spectrometer (Waters Corporation, U.K., Model: Alliance 2795, Q-TOF Micro-mass Mass spectrometer).

**2.2.5. Toxicity study.** The growth of locally sourced mung seeds (*Vigna radiata*) was measured under four conditions: control ( $\text{ddH}_2\text{O}$ ), untreated dye water ( $5\text{ ppm}$ ), treated  $\text{Al}_2\text{O}_3$  water, and treated ALOOH water. A total of  $250\text{ mL}$  of dye solution ( $5\text{ ppm}$  MB) was prepared in bulk. Notably,  $\text{Al}_2\text{O}_3$

and ALOOH were each used to degrade  $250\text{ mL}$  of  $5\text{ ppm}$  MB, each producing  $250\text{ mL}$  of water treated by  $\text{Al}_2\text{O}_3$  and  $250\text{ mL}$  of water treated by ALOOH. The water that was treated in bulk was used for toxicity assessment. Ten seeds were placed in each Petri dish, and  $15\text{ mL}$  of the respective solution was added daily. The Petri dishes were placed in a dark area and observed for 14 days.

The following equation was used to determine the germination index ( $G_i$ ):

$$G_i = \frac{G}{G_0} \times \frac{L}{L_0} (100) \quad (2)$$

where  $G$  and  $L$  are germination and root length, respectively, in the treated solution and  $G_0$  and  $L_0$  are germination and root length, respectively, in a  $100\%$   $\text{ddH}_2\text{O}$  control solution.<sup>26</sup>

**2.2.6. Reusability study.** The reusability of both particles was evaluated in three main cycles in neutral pH, under visible light conditions. In the first cycle, after the initial degradation of MB, the particles were retrieved and dried. These particles were then measured out again, and a degradation cycle was initiated. The quantities of the particles and solution were kept consistent with  $50\text{ mg}$  of particles in  $25\text{ mL}$  of solution.

## 3. Result and discussion

### 3.1. Synthesis and characterization

FTIR and XRD were used to characterize the prepared particles. Fig. 2(a) presents the contrast between the FTIR spectrum of the synthesized  $\text{Al}_2\text{O}_3$  (black) and ALOOH (red). Both samples showed peaks in the fingerprint region that can be attributed to bending and stretching vibrations, which are characteristics shared by organic compounds. Impurities in the waste aluminium foil that was used for the synthesis may be responsible for the presence of organic compounds.<sup>27</sup> Peaks seen for  $\text{Al}_2\text{O}_3$  at  $482$ ,  $532$ ,  $952$ , and  $1385\text{ cm}^{-1}$  are the bending and stretching of the Al-O interactions.<sup>27,28</sup> For ALOOH, these peaks were seen at  $481$ ,  $581$ ,  $713$ ,  $973$ , and  $1395\text{ cm}^{-1}$ . In addition to representing the Al-O interactions, peaks at lower wavelengths also signify a pseudo-boehmite structure. Specifically, this incomplete crystalline boehmite structure is seen in both molecules and contains aluminium-based matrix materials.<sup>29,30</sup> A band in  $\text{Al}_2\text{O}_3$  is also observed at  $656\text{ cm}^{-1}$ , which is correlated with the Al-O-Al interactions within the molecule.<sup>27</sup> The peaks in  $\text{Al}_2\text{O}_3$  at  $1556\text{ cm}^{-1}$  and in ALOOH at  $1668\text{ cm}^{-1}$  can be attributed to a *cis*-double bond in the molecule.<sup>27</sup> Bands are also seen at  $726\text{ cm}^{-1}$ , which are correlated with the tetrahedral structures found in  $\text{Al}_2\text{O}_3$ .<sup>26</sup> The broad peaks seen in samples around  $3302\text{ cm}^{-1}$  ( $\text{Al}_2\text{O}_3$ ) and  $3423\text{ cm}^{-1}$  (ALOOH) represent the vibration of the Al-O-H bonds in the sample.<sup>31</sup>

XRD is used to interpret the structures of atoms found inside a sample and can determine whether a sample is crystalline or amorphous. The XRD results in Fig. 2(b) were produced by  $\text{Al}_2\text{O}_3$  and ALOOH. For  $\text{Al}_2\text{O}_3$ , peaks were observed at  $\theta = 13.17^\circ$ ,  $18.84^\circ$ ,  $20.33^\circ$ ,  $27.95^\circ$ ,  $33.13^\circ$ ,  $40.66^\circ$ ,  $49.24^\circ$ ,  $53.33^\circ$ ,  $63.99^\circ$ , and  $70.98^\circ$ . These results were comparable with the XRD results for pure  $\text{Al}_2\text{O}_3$  given in the JCPDS.<sup>32</sup> It can be



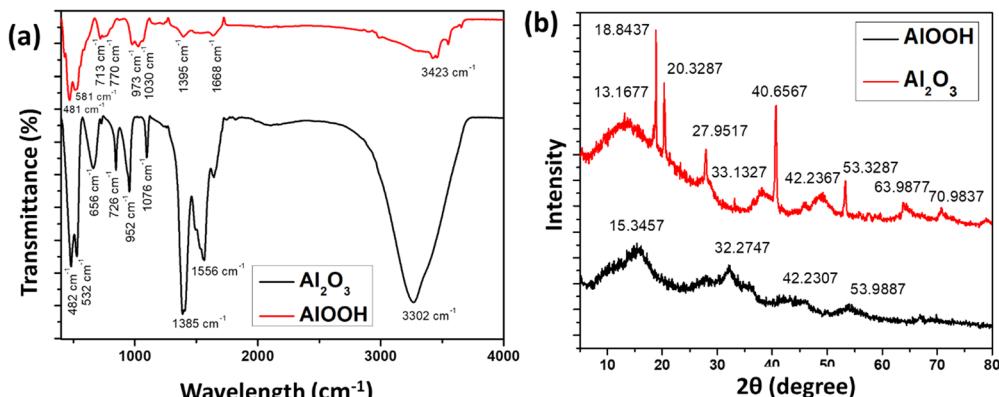


Fig. 2 FTIR (a) of  $\text{Al}_2\text{O}_3$  (black) and  $\text{AlOOH}$  (red). XRD (b) of  $\text{Al}_2\text{O}_3$  (red) and  $\text{AlOOH}$  (black).

inferred that peaks  $2\theta = 27.95^\circ$ ,  $49.24^\circ$ ,  $53.33^\circ$ , and  $63.99^\circ$  are caused by the presence of  $\text{NaCl}$ . These  $\text{NaCl}$  peaks are further confirmed in the literature.<sup>33</sup> It can also be inferred that the peaks at  $2\theta = 27.95^\circ$ ,  $40.66^\circ$ ,  $49.24^\circ$ , and  $70.98^\circ$  are caused by the presence of  $\text{Al}_2\text{O}_3$ . A sharp peak implies that the cell arrangement of the sample is crystalline.<sup>32,34</sup> Peaks for  $\text{AlOOH}$  were observed at  $2\theta = 15.35^\circ$ ,  $32.27^\circ$ ,  $43.23^\circ$ , and  $53.99^\circ$ . Saravanan *et al.* (2023) found the XRD pattern for  $\text{AlOOH}$  at  $2\theta = 27.4^\circ$ ,  $38.3^\circ$ ,  $48.8^\circ$  and  $64.9^\circ$ .<sup>25,35</sup> The broad width of the peaks suggests that  $\text{AlOOH}$  consists of nanocrystals; however, the peaks are not sharp suggesting that the molecule consists of an orthorhombic structure.<sup>25,36</sup> The peaks observed in the  $\text{AlOOH}$  are in correspondence with the literature, but some negative shift in the  $2\theta$  was found. This may be due to the  $\text{NaCl}$  present in the sample.

The absorbances of  $\text{Al}_2\text{O}_3$  (10 mg in 2 mL) and  $\text{AlOOH}$  (10 mg in 2 mL) in water were also recorded using UV-vis spectrophotometry, as can be seen in Fig. S1 (ESI†). It was observed that the absorption peaks of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  in  $\text{ddH}_2\text{O}$  match the literature results.<sup>37,38</sup> The band gaps of the  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  samples were calculated by performing a UV-vis DRS study. It is reported that the band gap changes with the crystalline nature of  $\text{Al}_2\text{O}_3$  and can be modulated when combined with other materials; however, if the calcination process is not complete, the surface hydroxyl groups can reduce the band gap, allowing it to act as a photocatalyst.<sup>39–41</sup> Edalati *et al.* reduced the  $\text{Al}_2\text{O}_3$  band gap to less than 3 eV using a high pressure torsion (HPT) method.<sup>42</sup>

Kusuma *et al.* explored the photocatalytic properties of  $\text{Al}_2\text{O}_3$  where  $\text{Al}_2\text{O}_3$  nanoparticles exhibited an energy band gap of 4.46 eV; these particles were used for MB degradation.<sup>43</sup> Additionally, Nduni *et al.* reported that  $\text{Al}_2\text{O}_3$  particles prepared using a green approach from waste aluminium foil have a band gap of 5.25 eV.<sup>27</sup> It is evident that  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  can degrade dye in the presence of light; however, they are unable to perform this function in the dark. This suggests that these compounds can act as photocatalysts. To further support these findings, the band gap energy was determined using a Tauc plot.<sup>38</sup> Fig. S2 (ESI†) depicts  $(\alpha h\nu)^2$  vs. energy (eV) for  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$ . It was determined that  $\text{Al}_2\text{O}_3$  has a band gap of 4.29 eV and  $\text{AlOOH}$

has a band gap of 5.50 eV. The band gap of  $\text{Al}_2\text{O}_3$  at 4.29 eV proves that this particle possibly has a photocatalytic nature.

The TGA results for  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  can be seen in Fig. S3(a) and (b) (ESI†). A multi-step decomposition for both particles is observed. In  $\text{Al}_2\text{O}_3$  (Fig. S3(a), ESI†), there is a 38.16% (2.15 mg) initial loss in weight (up to 100 °C), indicating dehydration. Then, there is a 20.13% loss in mass (1.14 mg) around 300 °C indicating decomposition. The decomposition that has occurred at this temperature matched the results observed by Gondal *et al.*<sup>44</sup> In total, there was about a 70.97% loss in mass when the temperature reached 900 °C.  $\text{AlOOH}$  (Fig. S3(b), ESI†) also follows a similar multistep decomposition. Initially, 22.29% of the total mass was lost between 50 and 100 °C, suggesting dehydration. Then, around 250–300 °C, there was an 11.77% mass loss due to decomposition. There was a 52.633% mass loss when the temperature reached 900 °C.

The heat flow in the  $\text{Al}_2\text{O}_3$  sample was detected using DSC measurement. Endothermic dips are seen at 100 °C and 300 °C, which is the same temperature range at which decomposition occurs. This implies that the sample is melting at these temperatures. DSC peaks are seen at 200 °C and 700 °C suggesting some crystallization in the sample. The heat flow in the  $\text{AlOOH}$  sample was also detected using DSC measurement. Endothermic dips are seen around 50–100 °C and 250–300 °C, which is the same temperature range at which decomposition occurs. This implies that the sample is melting at these temperatures. A DSC peak is seen at 275 °C revealing some crystallization in the sample.

The FE-SEM results in Fig. 3 confirm the surface morphology of the particles. It was observed that  $\text{Al}_2\text{O}_3$  has a jagged and rough surface that consists of different size pores, as can be seen in Fig. 3(a) and (b). The surface of this particle is more heterogeneous and larger than the that of the  $\text{AlOOH}$  particle. The  $\text{AlOOH}$  particle has a much smaller surface area than  $\text{Al}_2\text{O}_3$  (Fig. 3(c) and (d)). These nanoparticles also contain various-sized cavities and pores, making the particle look slightly rugged.

The results for the mapping and EDX of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  are presented in Fig. 4. The elements that were observed in the mapping of  $\text{Al}_2\text{O}_3$  mainly consist of Al and O atoms

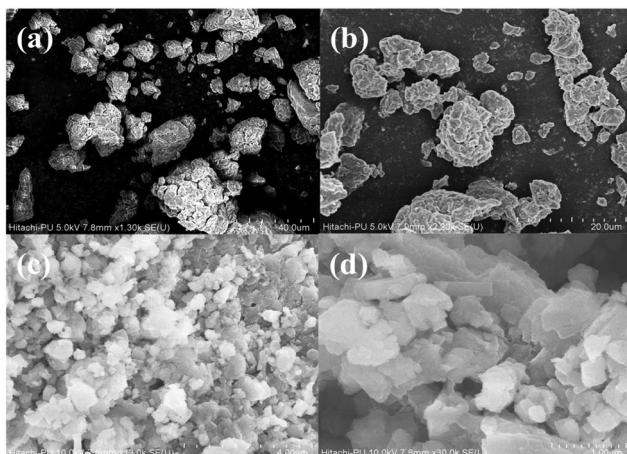


Fig. 3 FE-SEM of  $\text{Al}_2\text{O}_3$  (a) and (b) particles at scales of 40.0  $\mu\text{m}$  and 20.0  $\mu\text{m}$ , respectively. FESEM of AlOOH (c) and (d) particles at scales of 4.0  $\mu\text{m}$  and 1.0  $\mu\text{m}$ , respectively.

(Fig. 4(a)–(c)). There is a smaller amount of Na and Cl atoms, which can be attributed to the HCl in the synthesis of the particles (Fig. 4(d)). These can be constituted as impurities in the sample. The elements that were observed in the mapping and EDX of AlOOH also mainly consist of Al and O with a much smaller amount of Cl and Na (Fig. 4(e)–(h)). Overall, the mapping verifies that Al and O atoms are distributed homogeneously on the surface of both particles. The gold

(Au) present in the EDX mapping is due to the Au coating that was applied to  $\text{Al}_2\text{O}_3$  and AlOOH during the FESEM process.

The zeta-sizer results are presented in Table 1 and Fig. S4–S6 (ESI†). The hydrodynamic diameter and polydispersity index of the synthesized  $\text{Al}_2\text{O}_3$  and AlOOH were calculated at pH 5, 7 and 9. This study shows that at pH 5,  $\text{Al}_2\text{O}_3$  particles have a hydrodynamic diameter of around 3257 nm, which decreases with pH change to 1133 nm and 856.9 nm at pH 7 and 9, respectively. For AlOOH particles, the hydrodynamic diameter also changes with the change in pH value. Overall, the PDI values for  $\text{Al}_2\text{O}_3$  and AlOOH particles are still less than 52%. The zeta-potential value for  $\text{Al}_2\text{O}_3$  was 11 mV at 5 pH, and by changing the pH to 7 and 9, the zeta potential changed to –15.7 mV and –26.9 mV, respectively. At pH 5 for AlOOH, the zeta potential was –20.7 mV, and it changed to 4.3 mV and –17.0 mV at pH 7 and 9, respectively.

### 3.2. Degradation of dye under pH 7, pH 9, and pH 5

Fig. 5 shows a comprehensive analysis of the degradation of MB under different conditions using  $\text{Al}_2\text{O}_3$  and AlOOH as catalysts, in the dark and under visible light at pH 7. Fig. 5(a) depicts the MB solution with  $\text{Al}_2\text{O}_3$  in the dark observed for 240 minutes. This reaction was performed at pH 7. Initially, the concentration of the solution decreased, followed by stabilization. Fig. 5(b) depicts the MB solution with AlOOH in the dark observed for 240 minutes at pH 7. Like  $\text{Al}_2\text{O}_3$ , the concentration of the solution initially decreased and stabilized.

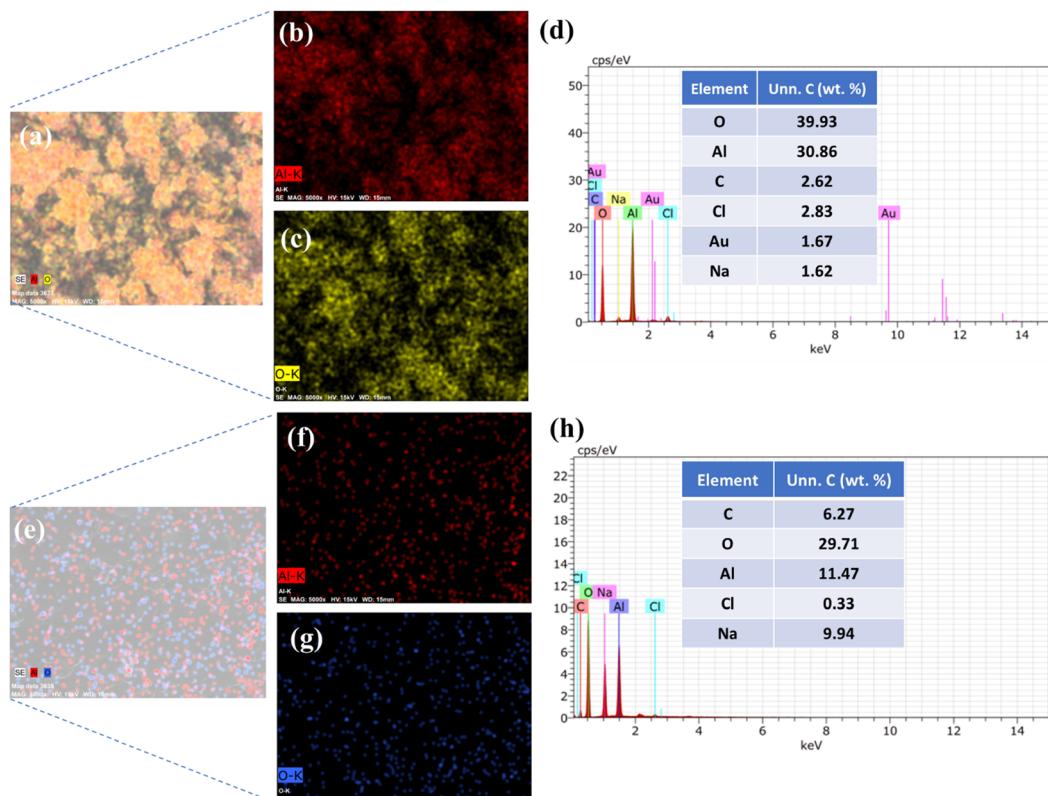


Fig. 4 Mapping of  $\text{Al}_2\text{O}_3$  (a)–(c) and EDX of  $\text{Al}_2\text{O}_3$  (d). Mapping of AlOOH (e)–(g) and EDX of AlOOH (h).



**Table 1** Hydrodynamic diameter, polydispersity index, and zeta potential of  $\text{Al}_2\text{O}_3$  and ALOOH

pH	Sample	Hydrodynamic diameter ( $D_h$ ) (nm)	Polydispersity index (PDI) (%)	Zeta potential (mV)
5	$\text{Al}_2\text{O}_3$	3257	47.8	11
	ALOOH	1803.4	37.0	-20.7
7	$\text{Al}_2\text{O}_3$	1133	52.6	-15.7
	ALOOH	3513	38.9	4.3
9	$\text{Al}_2\text{O}_3$	856.9	31.4	-26.9
	ALOOH	806.5	8.8	-17.0

The range of concentration decrease was significantly greater with  $\text{Al}_2\text{O}_3$  compared to ALOOH in the absence of light, which also signifies the adsorption of dye on the particles in the dark. Fig. 5(c) depicts the MB solution with  $\text{Al}_2\text{O}_3$  in visible light observed for 240 minutes at pH 7. The concentration of the solution steadily decreased for 240 minutes, and it eventually became clear. This demonstrates the photocatalytic degradation of MB under visible light by  $\text{Al}_2\text{O}_3$ . Fig. 5(d) depicts the MB solution with ALOOH in visible light observed for 240 minutes, at pH 7. The concentration of the solution steadily decreased for 240 minutes. Fig. 5(e) shows the time-dependent concentration decrease in dye degradation with irradiation time.

When the reactions were performed in the dark under neutral pH conditions, there was a 17.60% and 6.64% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  and ALOOH, respectively. In contrast, when the reactions were performed in visible light, under neutral conditions, a 99.29% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  and a 75.98% degradation in the MB solution with ALOOH were observed. Under dark conditions, the concentration initially dropped within the first 30–60 minutes before slowly becoming constant. This can be attributed to the first adsorption of the MB into the particles, which was then released back into the solution, causing the concentration to slightly fluctuate. The degradation efficiency of MB ( $C_t/C_0$ ) vs. time can also be seen in Fig. 5(e), which is the average of trial 1 and trial 2. This indicates that ALOOH also facilitates the photocatalytic degradation of MB under visible light, although possibly less efficiently than  $\text{Al}_2\text{O}_3$ . The same experiment was performed at pH 9, and accordingly, Fig. 6(a) represents the MB solution with  $\text{Al}_2\text{O}_3$  in the dark observed for 240 minutes. The concentration of this solution drastically decreased for 60 minutes and stabilized with some inconsistencies. Fig. 6(b) shows the MB solution with ALOOH in the dark observed for 240 minutes at pH 9. The concentration of this solution drastically decreased for 90 minutes, after which it again became stable with slight oscillation.

Fig. 6(c) depicts the MB solution with  $\text{Al}_2\text{O}_3$  in visible light observed for 240 minutes at pH 9. The concentration of the solution consistently decreased for 240 minutes, until it reached 0.21 a.u. Fig. 6(d) represents the MB solution with ALOOH in visible light observed for 240 minutes at pH 9. The concentration of the solution consistently decreased for 240 minutes, until it reached 0.18 a.u.

When the reactions were performed in the dark, under basic conditions, there was a 37.88% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  and a 37.70% degradation in the MB solution with

ALOOH. Despite their similar degradation efficiencies, ALOOH initially adsorbed MB more effectively than  $\text{Al}_2\text{O}_3$  under basic conditions in the dark. There was a larger range of adsorption in ALOOH than in  $\text{Al}_2\text{O}_3$  in the dark. When the reactions were performed in visible light, under basic conditions, there was an 82.77% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  and an 84.58% degradation in the MB solution with ALOOH. Under dark conditions, the concentration was seen to drastically drop within 60–90 minutes before showing slight inconsistencies with slight upward and downward variations. The degradation efficiency of MB vs. time can also be seen in Fig. 6(e), which is the average of the two trials. It was observed that under basic conditions in the dark, ALOOH adsorbs MB more effectively than  $\text{Al}_2\text{O}_3$ . After the initial concentration change in MB, the concentrations of both solutions stabilize with slight variations.

Another experiment was conducted to evaluate the photocatalytic efficiency of the catalysts under acidic conditions. This reaction was performed at pH 5. Fig. 7(a) and (b) represent the MB solution with  $\text{Al}_2\text{O}_3$  and ALOOH, respectively, in the dark observed for 240 minutes. The concentration of the dye decreased for 30–60 minutes, but overall, the concentration of MB remained consistent throughout the 240 minutes. Fig. 7(c) and (d) depict the MB solution with  $\text{Al}_2\text{O}_3$  and ALOOH, respectively, in visible light observed for 240 minutes. With both particles, the concentration of MB consistently decreased for 240 minutes, until it reached a clear solution.

When the reactions were performed in the dark, under acidic conditions, a 12.34% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  was observed. There was a 33.22% degradation in the MB solution with ALOOH. ALOOH and  $\text{Al}_2\text{O}_3$  were equally effective at adsorbing MB in the dark. When this reaction was performed in visible light, there was a 97.88% degradation in the MB solution with  $\text{Al}_2\text{O}_3$  and an 86.67% degradation in the MB solution with ALOOH. The dye was consistently degraded in visible light with  $\text{Al}_2\text{O}_3$  and ALOOH. The graph in Fig. 7(e) depicts the averages of the pairs of trials that were performed during the degradation reactions of MB. It is observed that  $\text{Al}_2\text{O}_3$  and ALOOH show similar patterns in acidic conditions. Under light,  $\text{Al}_2\text{O}_3$  and ALOOH are consistently decreasing; however,  $\text{Al}_2\text{O}_3$  is still more effective than ALOOH.

Overall, it can be observed that the MB solution was degraded most efficiently under neutral conditions with visible light with  $\text{Al}_2\text{O}_3$ , while it was degraded least efficiently under neutral conditions in the dark with ALOOH (Table 2). The range between the highest and lowest degradation efficiencies under all conditions is 92.65%. Specifically, under neutral conditions,  $\text{Al}_2\text{O}_3$  performed better than ALOOH in the dark and in the light. There was significantly more degradation in the light than in the dark. This proves that the dye is primarily adsorbed in the dark but degraded in the light.  $\text{Al}_2\text{O}_3$  and ALOOH performed better in the dark after the pH was increased from neutral to alkaline (7 to 9). However, in the light at pH 9, the efficiency of  $\text{Al}_2\text{O}_3$  decreased while the efficiency of ALOOH increased by 8.59%. At pH 9, ALOOH is proven to be a more effective photocatalyst than when used at pH 7. In the dark at pH 5, ALOOH was significantly more effective (20.88% more effective) at degrading MB compared to  $\text{Al}_2\text{O}_3$ .



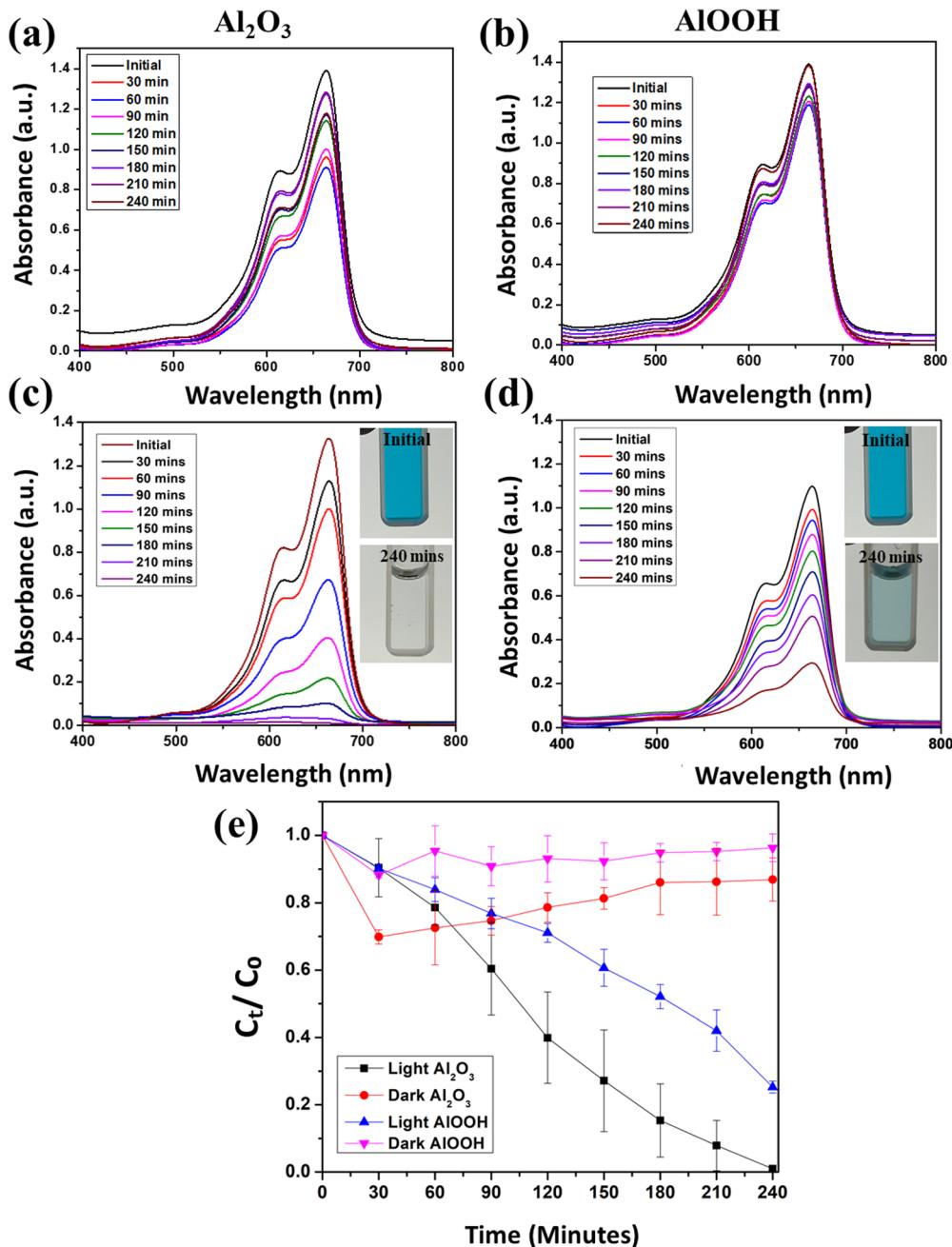


Fig. 5 UV-vis spectra of MB with Al<sub>2</sub>O<sub>3</sub> (a) and AlOOH (b) in the dark and Al<sub>2</sub>O<sub>3</sub> (c) and AlOOH (d) in the light at pH 7. Average degradation efficiency over time at pH 7 (e).

However, in the light under acidic conditions, Al<sub>2</sub>O<sub>3</sub> was 11.21% more effective than AlOOH at degrading MB.

When only the dark conditions are considered at all pH values, it is observed that Al<sub>2</sub>O<sub>3</sub> is least effective at pH 5 and most effective at pH 9 and AlOOH is least effective at pH 7 and most effective at pH 9. The range between the highest and lowest degradation efficiencies in the dark under all pH values is 31.24%. When only visible light conditions are considered at all pH values, it is observed that Al<sub>2</sub>O<sub>3</sub> is consistently more effective than AlOOH for the photocatalytic degradation of MB. At pH 7, Al<sub>2</sub>O<sub>3</sub> is the most effective, while AlOOH is the least

effective. Nevertheless, it was discovered that the efficiency of AlOOH was improved by increasing and decreasing the pH. The range between the highest and lowest degradation efficiencies in visible light under all pH values is 23.30% (Table 2).

In order to determine the kinetics of dye degradation, first-order kinetics were applied:  $\ln(C_t/C_0) = -kt$ , where  $C_t$  and  $C_0$  are the final concentration at that particular point of time (min) and the initial concentration of the MB dye, respectively, and  $k$  is the rate constant. Fig. S7–S9 (ESI<sup>†</sup>) depict plots of  $\ln(C_t/C_0)$  vs. time (min) intervals, showing a linear correlation obtained by plotting the degradation of MB and suggesting that the

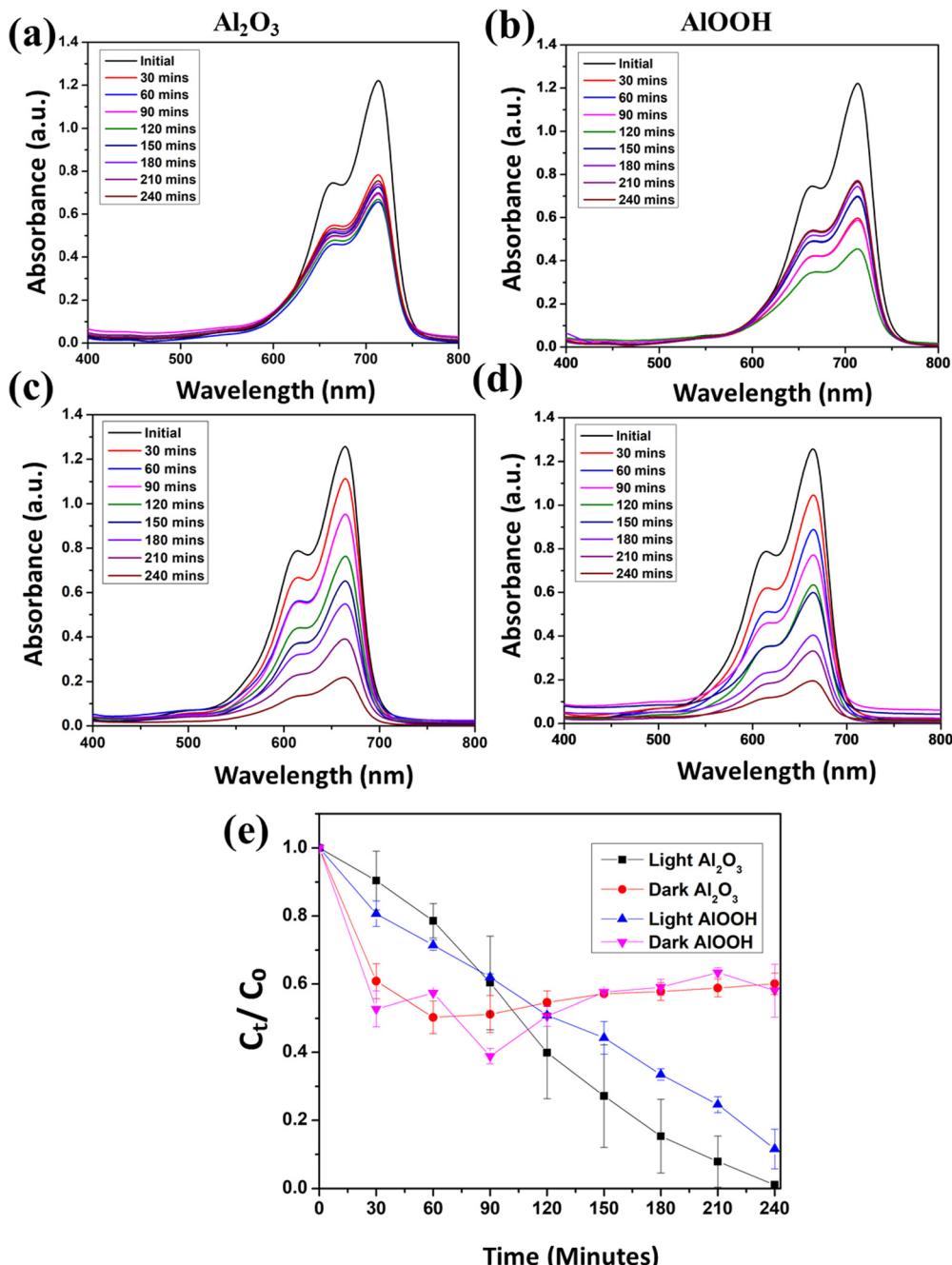


Fig. 6 UV-vis spectra of MB with  $\text{Al}_2\text{O}_3$  (a) and  $\text{AlOOH}$  (b) in the dark and  $\text{Al}_2\text{O}_3$  (c) and  $\text{AlOOH}$  (d) in the light at pH 9. Average degradation efficiency over time at pH 9 (e).

removal of MB dye using  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  followed pseudo-first-order kinetics.

Fig. 8(a)–(e) illustrate the behavior of the MB solution in dark and light, with and without photocatalysts. The initial MB solution is shown in Fig. 8(a). After 240 minutes of light exposure, the solution becomes colorless in the presence of  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  (Fig. 8(b) and (c)). Conversely, under dark conditions, the solution remains colored with  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$  (Fig. 8(d) and (e)). This picture clearly shows the effect of the catalyst in the absence and presence of light, which also

confirmed that in the presence of light photodegradation occurred.

To further check the formation of intermediates after the effective photocatalytic activity of  $\text{Al}_2\text{O}_3$ , a liquid chromatography–mass spectrometry (LC–MS) test was performed on the initial MB, MB with  $\text{Al}_2\text{O}_3$  in the dark and MB with  $\text{Al}_2\text{O}_3$  under light after 240 min. Fig. 8(f)–(h) show the full-range mass scan graph of pure MB without treatment, MB with  $\text{Al}_2\text{O}_3$  in the dark and in the presence of light, respectively. Pure and MB with  $\text{Al}_2\text{O}_3$  under dark conditions showed very similar results



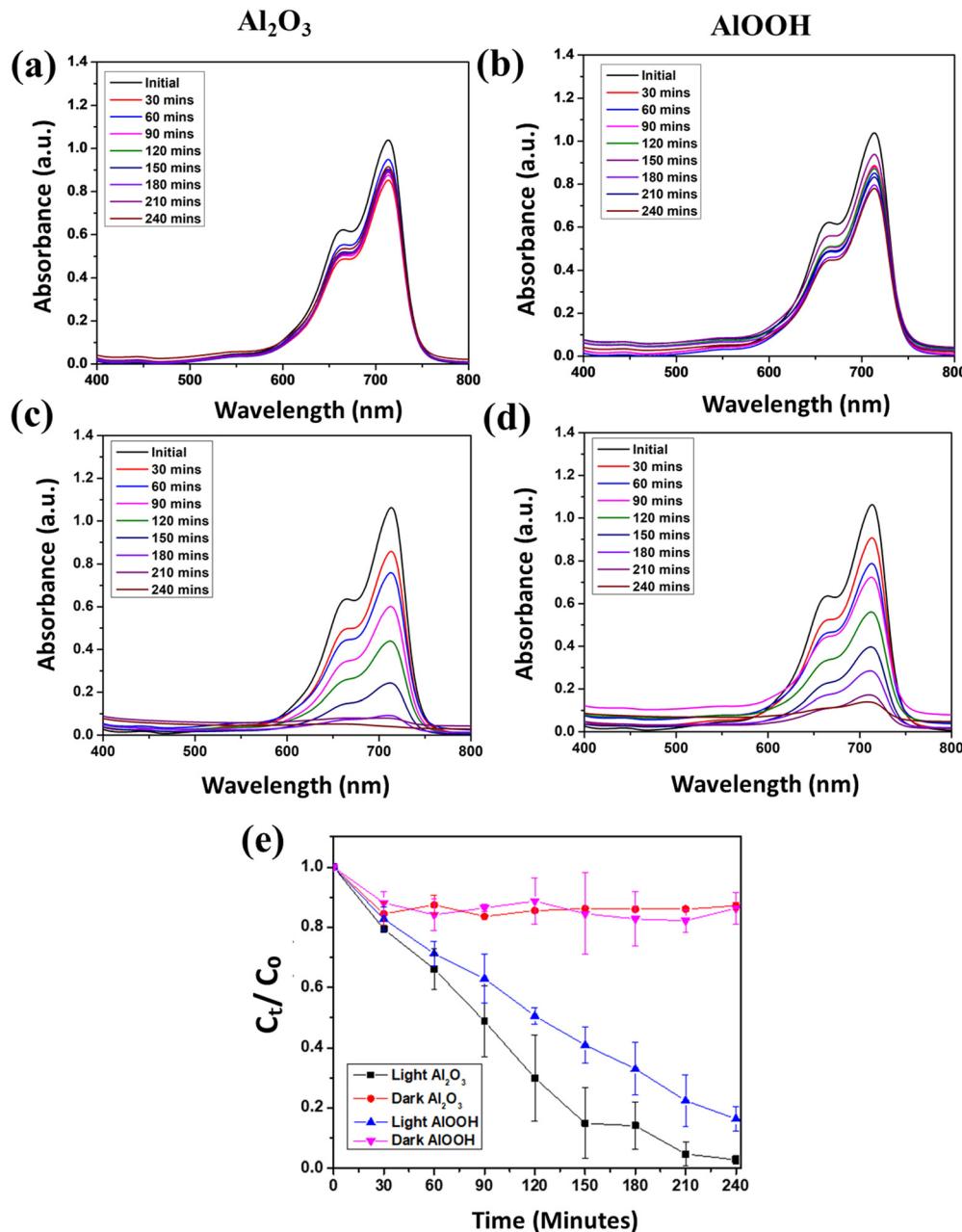


Fig. 7 UV-vis spectra of MB with  $\text{Al}_2\text{O}_3$  (a) and  $\text{AlOOH}$  (b) in the dark and  $\text{Al}_2\text{O}_3$  (c) and  $\text{AlOOH}$  (d) in the light at pH 5. Average degradation efficiency over time at pH 5 (e).

Table 2 Comparison between the degradation efficiencies at pH 7, pH 9, and pH 5 under dark and visible light conditions

	pH 5		pH 7		pH 9	
	$\text{Al}_2\text{O}_3$ (%)	$\text{AlOOH}$ (%)	$\text{Al}_2\text{O}_3$ (%)	$\text{AlOOH}$ (%)	$\text{Al}_2\text{O}_3$ (%)	$\text{AlOOH}$ (%)
Dark	12.34	33.22	17.60	6.64	37.88	37.70
Light	97.88	86.67	99.29	75.99	82.77	84.58

with peaks seen at approximately 270, 284 (largest peak), 285 and 286  $m/z$ . The maximum percentage peak at  $m/z = 284$  corresponds to the  $\text{M}^+$  molecular ion of methylene blue that

matches the reported literature value of the MB base peak value.<sup>45</sup> The identical results for pure and MB with  $\text{Al}_2\text{O}_3$  under dark conditions further showed that no photocatalytic intermediate products were formed. Conversely, when the experiment was performed in the light, the following peaks were observed: 72.93, 72.98, 88.98, 89.9, 96.96, 97.96, 112.95, 116.97, 130.00, 138.95, 158.00, 166.95, 179.98, 202.17, 207.98, 224.12, 241.92, 254.92, 267.94, and 281.89  $m/z$  with about 14 more distinguishable peaks. The significant increase in the number of peaks indicates that new products were formed as a result of the photodegradation of the original MB with  $\text{Al}_2\text{O}_3$ . Peaks seen at different  $m/z$  are consistent with the



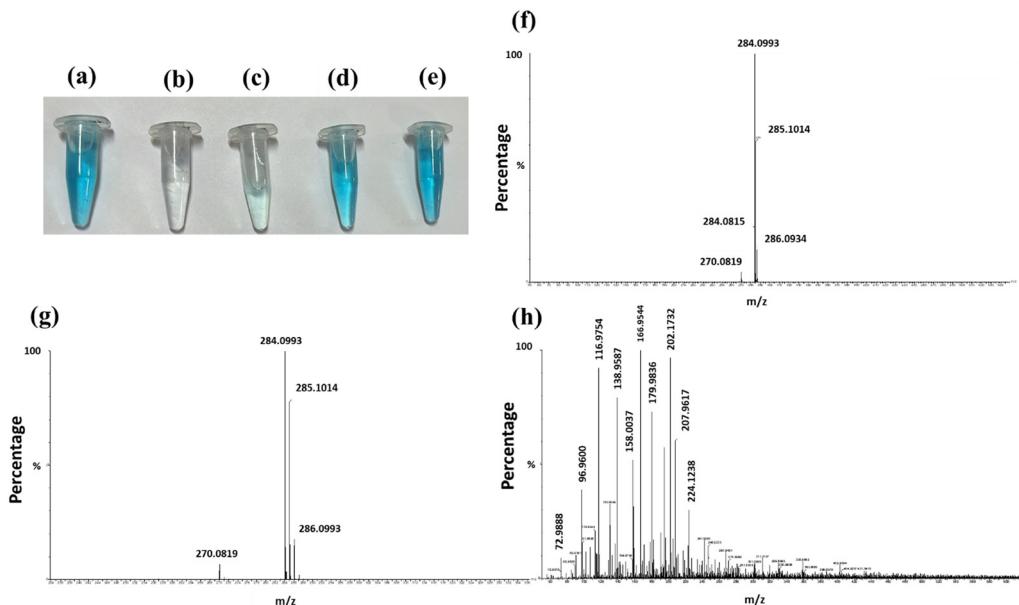


Fig. 8 Samples of MB: (a) initial, (b) with  $\text{Al}_2\text{O}_3$  in the light after 240 min, (c) with  $\text{AlOOH}$  in the light after 240 min, (d) with  $\text{Al}_2\text{O}_3$  in the dark after 240 min, and (e) with  $\text{AlOOH}$  in the dark 240 min. Mass spectra of MB: (f) initial, (g) 240 min in the dark, and (h) 240 min in the light.

Table 3 Number of seeds germinated and their respective lengths, used to calculate the germination index ( $G_i$ ) of mung seeds in control, MB,  $\text{Al}_2\text{O}_3$  and  $\text{AlOOH}$

Sample	#G	L (cm)	$G_i$
Control	18	76.25	100
MB	14	36.75	37.48634
$\text{Al}_2\text{O}_3$	18	34	44.59016
$\text{AlOOH}$	15	25.251	27.59563

possible mass spectra obtained for MB after degradation<sup>45</sup> (Fig. 8).

The presence of only three peaks in the initial LC-MS graphs indicates the primary molecular components of the sample. The lack of change in this sample after allowing the catalyst to interact with MB in the dark for 240 min suggests that the parent compound has not changed. However, when the sample was tested after exposure to light for 240 min, the appearance of different peak suggests that the original molecules have likely

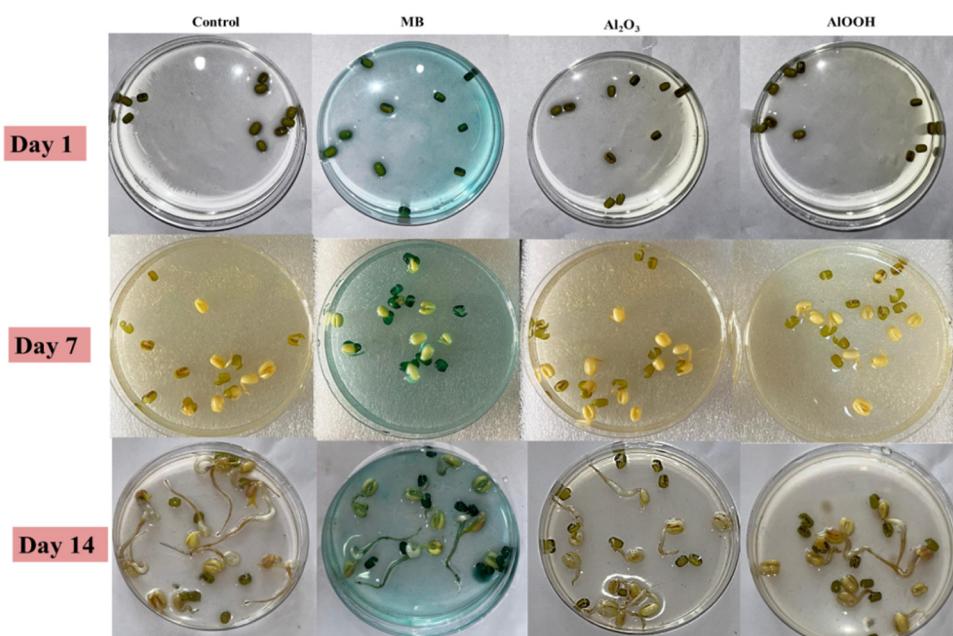


Fig. 9 Mung seed (*Vigna radiata*) growth in different conditions over two weeks (14 days).



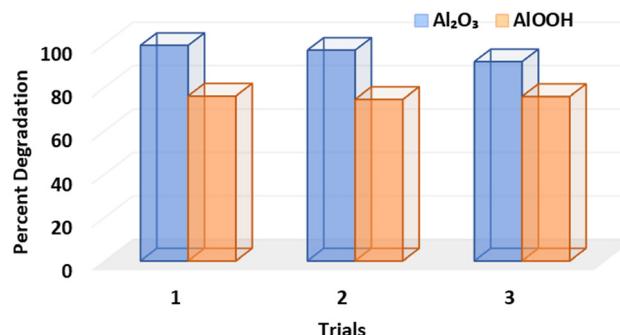


Fig. 10 The graphs of Al<sub>2</sub>O<sub>3</sub> and AlOOH after three cycles of using, drying, and reusing.

fragmented into smaller pieces, each with a different mass-to-charge ratio, leading to new peaks<sup>45,46</sup> (Fig. 8).

To further prove the photodegradation process of Al<sub>2</sub>O<sub>3</sub>, an experiment was also performed at a higher concentration of MB: 20 ppm. The results of the experiment in the dark and in the light are presented in Fig. S10 (ESI†). The results show that MB continues to degrade under light with Al<sub>2</sub>O<sub>3</sub>.

For the toxicity analysis, the mung seeds were incubated at room temperature for 14 days in the dark.<sup>47</sup> After incubation, the number of seeds that germinated (G) and the root length (L) were measured for seeds in each condition: ddH<sub>2</sub>O, 5 ppm MB, treated Al<sub>2</sub>O<sub>3</sub> water, and treated AlOOH water. The MB solution was treated (under visible light) with Al<sub>2</sub>O<sub>3</sub> and AlOOH in water in bulk reactions, proving that these particles can operate in larger-scale reactions.

From the results of the toxicity assessment, it was concluded that ddH<sub>2</sub>O produced the most germinated seeds followed by Al<sub>2</sub>O<sub>3</sub>, AlOOH, and MB (Table 3 and Fig. 9). However, despite having more germinated seeds, the germination index for AlOOH was lower than that of MB, as the seeds in MB solution grew longer. This increased the overall germination index.

Excellent results were seen in Al<sub>2</sub>O<sub>3</sub> and AlOOH particles when tested for reusability. This assessment was performed

under visible light conditions at pH 7. As can be seen in the initial graphs (Fig. 5), Al<sub>2</sub>O<sub>3</sub> was able to degrade 99.29% of the MB dye; in the second cycle, the degradation efficiency was 97.09%; and in the third cycle, the degradation efficiency was 91.79%. The degradation of AlOOH followed a similar pattern (Fig. 5), indicating a 75.99% degradation under light conditions. The second and third cycles then show a 74.47% and 75.78% degradation of MB, respectively (Fig. 10).

A comparative study was used to assess the degradation efficiency of various metal oxide nanoparticles in different dyes. Dyes including malachite green, brilliant crystal, and methylene blue were examined.<sup>15</sup> The findings of this comparison revealed that the synthesized Al<sub>2</sub>O<sub>3</sub> exhibited the comparable effectiveness in degrading the MB dye.

Table 4 provides a comprehensive summary of Al<sub>2</sub>O<sub>3</sub> and other metal oxides prepared by various methods that have been used for photocatalytic degradation under different light sources. As shown in the Table 4, the MB degradation efficiencies are comparable with the irradiation times in many studies. However, some results differ from the findings of this paper due to variations in the synthesis processes and the use of different light sources, such as sunlight and UV illumination. Unlike previous studies where Al<sub>2</sub>O<sub>3</sub> was prepared electrochemically<sup>48</sup> or from aluminium salts like aluminium nitrate and sodium hydroxide,<sup>49</sup> our novel study uses waste aluminium foil for a green synthesis of the particles. The Al<sub>2</sub>O<sub>3</sub> and AlOOH synthesized in this manner can serve not only as photocatalysts but also in various other industries and processes.

## 4. Conclusions

This study successfully demonstrated that compromised foil can be reused to synthesize Al<sub>2</sub>O<sub>3</sub> and AlOOH particles that are effective photocatalysts for dye degradation. This comprehensive research analyzed the particle structure, shape, bonds, composition, thermal stability, degradation efficiency, toxicity, and reusability. The particles were observed under neutral,

Table 4 Analysis of photocatalytic efficiencies of various metal oxides, dyes, times, and degradation percentages

Metal oxide NPs	Photocatalyst amount	Concentration of dye	Dye	Degradation time (min) (light source)	Degradation percent (%)	Ref.
Al <sub>2</sub> O <sub>3</sub> prepared by sol-gel method	10 mg	0.01 mM of MB	MB	240 (sunlight)	sol-gel-Al <sub>2</sub> O <sub>3</sub> : 85.0% γ-Al <sub>2</sub> O <sub>3</sub> : 91.6%	14
γ-Al <sub>2</sub> O <sub>3</sub> prepared by precipitation method						
Al <sub>2</sub> O <sub>3</sub>	100 mg	1.5 × 10 <sup>-5</sup> mol L <sup>-1</sup>	malachite green	360 (sunlight)	45%	49
γ-Al <sub>2</sub> O <sub>3</sub> NPs	60 mg	20 ppm	MB	60 (UV light)	96.4%	48
N-ZnO	50 mg	100 mL of MB 20 mg L <sup>-1</sup>	MB	80 (solar-simulated light)	95.3%	50
MnFe <sub>2</sub> O <sub>4</sub>	30 mg	10 ppm	MB	290 (UV light)	97%	51
Al <sub>2</sub> O <sub>3</sub> /SiO <sub>2</sub>	300 mg	—	malachite green	300 (sunlight)	85%	52
Al <sub>2</sub> O <sub>3</sub> /Fe <sub>2</sub> O <sub>3</sub>	200 mg	100 mL of MB solution (25 mg L <sup>-1</sup> )	MB	90 (visible light)	75.1%	53
Biosynthesized ZnO	20 mg	2.0 × 10 <sup>-5</sup> M	MB	420 (visible light)	98%	54
AlOOH	50 mg	5 ppm	MB	240 (visible light)	75.99%	Current
Al <sub>2</sub> O <sub>3</sub>	50 mg	5 ppm	MB	240 (visible light)	99.29%	Current



basic, and acidic conditions. The results convey that the MB degradation was most efficient in light for any pH considered. The experiments further revealed that in the light, the highest efficiency was observed when using  $\text{Al}_2\text{O}_3$  under neutral conditions, achieving a degradation efficiency of 99.29%. Additionally, the highest efficiency was achieved in the light with  $\text{AlOOH}$  under acidic conditions, with a degradation efficiency of 86.67%. It was also observed that the pH conditions affect the degradation of the dye solution in the light and dark. Moreover, it was observed that in the dark, the absorbance of the dye decreased initially and stabilized while there was a constant decrease in the absorbance of the dye in the light. This proved that MB dye is primarily absorbed in the dark but degraded in the light. The toxicity results further revealed that control,  $\text{Al}_2\text{O}_3$ ,  $\text{AlOOH}$ , and MB produced the most germinated seeds. The germination indexes of the solutions were observed in the following order: control,  $\text{Al}_2\text{O}_3$ , MB, and  $\text{AlOOH}$ . The toxicity assessment also proved that these particles could degrade MB in bulk reactions. Both particles also showed remarkable recyclability for at least three cycles.

## Data availability

Data supporting this study are included within the article and/or ESI.<sup>†</sup>

## Conflicts of interest

There are no conflicts to declare.

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