coatings

MDPI

Review

The Suitability of Photocatalyst Precursor Materials in Geopolymer Coating Applications: A Review

Liyana Jamaludin [1,2,*], Rafiza Abd Razak [1,3], Mohd Mustafa Al Bakri Abdullah [1,4,*], Petrica Vizureanu [5,6,*], Ana Bras [7], Thanongsak Imjai [8], Andrei Victor Sandu [5,9,10], Shayfull Zamree Abd Rahim [2] and Heah Cheng Yong [2]

1 Centre of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis (UniMAP), Perlis 01000, Malaysia
2 Faculty of Mechanical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), Perlis 01000, Malaysia
3 Faculty of Civil Engineering & Technology, Universiti Malaysia Perlis (UniMAP), Perlis 01000, Malaysia
4 Faculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), Perlis 01000, Malaysia
5 Faculty of Materials Science and Engineering, Gheorghe Asachi Technical University of Iasi, 41 D. Mangeron Street, 700050 Iasi, Romania
6 Technical Sciences Academy of Romania, Dacia Blvd 26, 030167 Bucharest, Romania
7 Built Environment and Sustainable Technologies (BEST) Research Institute, Liverpool John Moores University, Liverpool L3 2ET, UK
8 Center of Excellence in Sustainable Disaster Management, School of Engineering and Technology, Walailak University, Nakhonsithammarat 80161, Thailand
9 Department of Research, Development and Innovation, Romanian Inventors Forum, Str. Sf. P. Movila 3, 700089 Iasi, Romania
10 National Institute for Research and Development for Environmental Protection INCDPM, 294 Splaiul Independentei, 060031 Bucharest, Romania
* Correspondence: liyanajamaludin@unimap.edu.my (L.J.); mustafa_albakri@unimap.edu.my (M.M.A.B.A.); peviz2002@yahoo.com (P.V.)

check for updates

Citation: Jamaludin, L.; Razak, R.A.; Abdullah, M.M.A.B.; Vizureanu, P.; Bras, A.; Imjai, T.; Sandu, A.V.; Abd Rahim, S.Z.; Yong, H.C. The Suitability of Photocatalyst Precursor Materials in Geopolymer Coating Applications: A Review. *Coatings* **2022**, *12*, 1348. https://doi.org/10.3390/coatings12091348

Academic Editor: Csaba Balázsi

Received: 19 August 2022
Accepted: 12 September 2022
Published: 16 September 2022

Abstract: Today, the building and construction sector demands environmentally friendly and sustainable protective coatings using inorganic coating materials for safe, non-hazardous, and great performance. Many researchers have been working on sustainable solutions to protect concrete and metal infrastructures against corrosion and surface deterioration with the intention of introducing green alternatives to conventional coatings. This article presents a review of developments of geopolymer pastes doped with different types of photocatalyst precursors including factors affecting geopolymer properties for enhancing coating with photocatalytic performance. Photodegradation using geopolymer photocatalyst has great potential for resolving harmless substances and removing pollutants when energized with ultraviolet (UV) light. Although geopolymer is a potentially new material with great properties, there has been less research focusing on the development of this coating. This study demonstrated that geopolymer binders are ideal precursor support materials for the synthesis of photocatalytic materials, with a significant potential for optimizing their distinctive properties.

Keywords: coating; geopolymer; photocatalytic; photocatalyst precursor

1. Introduction

Geopolymers represent novel material types at the interface of glass, ceramics, and materials based on traditional inorganic bonds. Geopolymers utilize waste materials as source material and activate the materials with alkaline activators to act as binders. Metakaolin is categorised as an aluminosilicate material because it contains variable amounts of alumina and silica [1]. Geopolymers offer benefits due to their ease of synthesis and low emissions of greenhouse gases such as CO_2, SO_2, and NO_x [2,3]. Furthermore, because it uses minerals of geological origin, the procedure is relatively green and eco-friendly. The benefits of greenhouse gas reduction and waste management applications utilising geopolymer materials have drawn significant global attention to the sustainable development of technology for industrial waste utilisation in building construction.

Currently, the potential of a building to get dirty is a recurring problem, particularly when it has been subjected to high levels of pollution over an extended period of time [4]. Natural disasters and the collapse of historic structures release concrete dust, resulting in a poor state of a building's surface, such as a gritty and grey look. As a result, air pollution has become a significant environmental concern. Furthermore, the usage of volatile organic compounds (VOCs) in paints has been shown to degrade the ozone layer [5]. These harmful substances are known to have negative human health effects, and this issue has become an issue in the construction industry. Some VOCs deplete the ozone layer, while others are hazardous to human health. In response, the coatings industry has sought new environmentally friendly materials, limiting the use of epoxy and metals such as chromium and lead in pigments and minimising emissions of the most volatile organic compounds [6].

Geopolymer coating has received much interest as an alternative material for protective coatings on concrete surfaces and other ceramic applications [7,8]. The emission of concrete dust due to the destruction of historic structures and natural disasters is also a problem that must be addressed. The advent of geopolymer coatings with photocatalytic properties is advantageous for pollutant degradation, surface deterioration prevention, anti-corrosion, and concrete self-cleaning [9]. Several methods utilise photocatalysts for a variety of applications, including the degradation of organic pollutants in wastewater, air purification, and antibacterial activity [10–12]. ZnO, TiO_2, WO_3, and Fe_2O_3 are nanoparticle materials used as photocatalyst precursors to enhance the functionality of cementitious materials [3,13,14]. When exposed to ultraviolet (UV) light, photocatalysts degrade organic and inorganic contaminants into less hazardous forms. Due to their lack of toxicity, chemical stability, and high reactivity, titanium-oxide compounds have shown promising potential as photocatalysts for numerous important processes [15,16]. ZnO are significant choices for photocatalysts in photodegradation because they are inexpensive, nontoxic, and excellent at absorbing a large percentage of the sunlight spectrum [17–19]. Several studies also discussed the uses of other nanoparticle materials such as V_2O_5, Ag_3VO_4, $LaVO_4$, and $BiVO_4$, which are utilized as photocatalyst precursor. According to Sharifi et al., $BiVO_4$ possesses rapid photogenerated charge recombination, poor electron transfer kinetics, and low photogenerated hole oxidative power and is suitable for advanced water treatment [20]. Shan et al. [21] also studied the photocatalytic efficiency of $BiVO_4$ and identified effective discoloration under sunlight exposure. Meanwhile, Shafiq et al. [22] concentrated on the use of $LaVO_4$ as a photocatalyst material with excellent visibly driven deep oxidative desulfurization capacity for use in petroleum refineries. Zhang et al. [23] agreed that $LaVO_4$ is effective as a photocatalyst for the decomposition of wastewater. Qi et al. [24] found V_2O_5-WO_3/TiO_2 to be a prominent catalyst for synthesizing NO_x. Geopolymers are eco-friendly and can easily incorporate with photocatalyst precursor materials such as ZnO and TiO_2, making them an excellent choice for construction applications, especially as coating material [25–29]. With exposure to UV-visible light, geopolymers containing photocatalyst precursor materials can oxidise and degrade surface contaminants on a building or road, allowing the products to be readily removed by rain, cleaning, or washing [9].

This review concentrates on prior research on photocatalyst precursor materials' potential to be utilized for the degradation of pollutants in wastewater, coating surfaces with Ordinary Portland Cement (OPC) self-cleaning concrete, the cleaning of air, and reducing VOC pollution when exposed to UV radiation from the sun. This paper also reviews the research on geopolymerization processes for coating compared with conventional coating. In addition, this research examines the use of geopolymer as an alternative coating doped nanoparticles photocatalyst precursor material. Geopolymer paste has the potential to be employed in the building sector as coating materials with improved physical and mechanical qualities. The geopolymer characteristics were enhanced by the addition of photocatalyst precursor components for coating applications. The current knowledge gaps for additional study have been identified based on substantial reviews. Nanoparticle photocatalysts were discovered to be commonly employed in traditional concrete and OPC for self-cleaning applications and degradation of dye in wastewater and air pollutants. On the other hand, most researchers identified that the photocatalytic effect typically requires complex methods such as sol gel and hydrothermal to achieve high photocatalyst performance; however, the geopolymer technology simplifies the whole procedure and utilizes inorganic materials, therefore addressing environmental concerns. Depending on the specifics of nanomaterials due to their strong reactivity and fine particle size, the use of nanoparticles in cement-based products attracted interest as well as unique functional properties. The use of geopolymer materials for coating applications, however, is still not fully investigated even though combinations of geopolymer materials and nanoparticles materials such as ZnO and TiO_2 have been proven as great alternatives for building applications, particularly as coating materials. The next section discusses the advantages of incorporating photocatalysts into geopolymer materials as well as the mechanism of photocatalytic reaction in the photodegradation process.

2. Geopolymer Aluminosilicate Materials

Geopolymers result from the interactions of inorganic elements like coal fly ash and incinerator ash, slags such granulated blast (steel) or furnace (iron) slag, and clays like metakaolin or calcined clay [30,31] with an alkaline activator. Geopolymers focus on utilizing waste products to create value. Other industrial wastes included glass, melt-quenched aluminosilicates, natural minerals such as kaolinite and natural zeolite, volcanic ash, and mine tailing, waste ceramics, and catalyst residues, as well as mixtures of these materials [9,32]. Fly ash and metakaolin are the most frequent aluminosilicates or raw materials employed by researchers to construct traditional geopolymer adsorbents. Geopolymers incorporate waste materials as source materials and an alkaline activator to serve as a binder. Commonly, the alkaline liquid utilised in geopolymerization is a mixture of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) [33,34]. Geopolymerization is a heterogeneous chemical reaction involving solid aluminosilicate oxides and alkali metal silicate solutions under very alkaline conditions and low temperatures that produces amorphous to semicrystalline polymeric structures composed of Si-O-Al and Si-O-Si bonds [35,36]. Geopolymerization entails a very rapid chemical reaction under alkaline circumstances with Si and Al minerals, resulting in a three-dimensional polymeric chain and ring structure composed of Si-O-Al-O bond [37].

Kaolin has high concentrations of SiO_2 and AlO_3 depending on the place of extraction. Kaolin is then extracted and subjected to the calcination process, which seeks to produce a material with pozzolanic features and high reactivity. Metakaolin ($Al_2Si_2O_7$) is made from kaolin clay. An amorphous kaolinite produced by treatment at 500 to 800 °C was used to convert kaolin to metakaolin [38,39]. The Al (VI) in kaolinite is converted into Al (IV) and Al (V) in this process to generate amorphous aluminium silicate. Spinel, mullite, and other crystals are formed when a high temperature is maintained constantly throughout the calcination process. Al (IV) and Al (V) will be changed into Al (VI) through this procedure [40]. In strong alkali solutions, metakaolin dissolves and releases Al and Si rapidly, producing geopolymer, zeolite, and other compounds depending on the reaction environment [41]. Metakaolin has substantially greater activity than kaolin in the same environment, which expands the application range of metakaolin, especially as geopolymer material.

Fly ash is a solid fine residue formed of particles expelled from the boilers of coal-fired power plants with flue gases [37,42,43]. Fly ash is used in the development of geopolymers because of its naturally high concentrations of SiO_2 and Al_2O_3; low SiO_2 and Al_2O_3 content is insufficient for alkali activation [44].

Slag is a by-product of the production of wrought iron and steel. As by-products of metallurgical operations or incineration processes, many slags are formed. In slag-blended systems, the geopolymerization reaction rate rises with increasing slag and activator concentrations [45]. Table 1 summarises the most recent published research on aluminosilicate materials. The table includes the aluminosilicate materials and the research findings. This summary shows that researchers are focusing more on using geopolymer aluminosilicate materials for concrete and cement applications and less on using the geopolymer materials for coating applications.

Table 1. Aluminosilicate materials used in the recent research on geopolymers.

Author	Aluminosilicate Material	Finding
Yusuf G. Adewuyi [37]	Class F Fly Ash	Elimination of trace noxious heavy metals in aqueous environment. The geopolymer adsorbent as a substance is recyclable since it can be synthesized by leveraging abundant waste materials.
Rafik Abbas et al. [46]	Kaolin	Used to produce geopolymer concrete as it does not require energy for pretreatment and contains high alumina silicate.
Abdulrahman et al. [47]	Metakaolin	Metakaolin geopolymer with different mix design for producing geopolymer concrete.
Ionescu et al. [48]	Slag	Steel slag or blast furnace slag in the production of geopolymer for construction building materials

3. Photocatalyst

3.1. Photocatalyst Mechanism

Volatile organic compounds (VOCs) are gases emitted by some solids and liquids. VOCs comprise an extensive variety of chemicals that may have short- and long-term adverse health effects. When compared with other technologies, photocatalysis are rapidly evolving and gaining popularity due to their numerous advantages, including low cost and great efficiency. The photocatalytic process is a potential method for eliminating volatile organic compounds (VOCs) from pollutant environments. Photocatalysis technology is a viable solution to environmental degradation in the construction and industrial sectors. Photocatalysis is the process that happens when a light source interacts with the surface of a substance. Photocatalytic reaction attributed to improved (UV) ray energy to oxidise organic compounds and degrade other contaminants to a less hazardous state when exposed to sunlight [49,50]. Photocatalytic response occurs when a surface is exposed to UV radiation from the sun. A photocatalyst is a substrate that absorbs light and functions as a catalyst for chemical processes [50]. Photocatalysis is a reaction that occurs when a semiconducting substance is exposed to light and generates an electron–hole pair [50–53]. The band gap is the energy difference between the valence bands (VB) and conduction bands (CB). The band gap energy of a semiconductor is the amount of energy required to shift an electron from the valence band to the conduction band [54]. As shown in Figure 1, the different types of material evaluate different band gap energy (Eg), which is other metal or conductor; Eg < 1.0 eV, semiconductors; Eg < 1.5–3.0 eV and insulators; Eg > 5 eV [50].

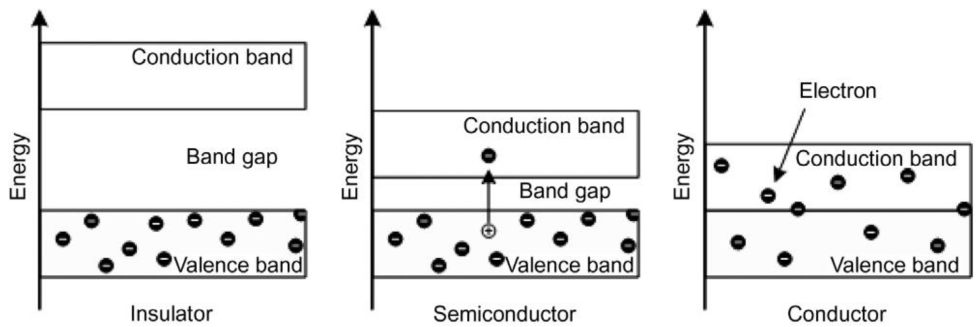


Figure 1. Band gap energy for different type of materials [50].

On exposure to light with energy equivalent to or more than its band gap energy, the semiconductor photocatalyst produces an oxidative and a reductive entity [55]. Figure 2 presents the valence and conduction bands, referred to as holes and electrons, respectively. Via a sufficient band gap, electrons move from the valence band to the conduction band in the presence of ultraviolet light [14].

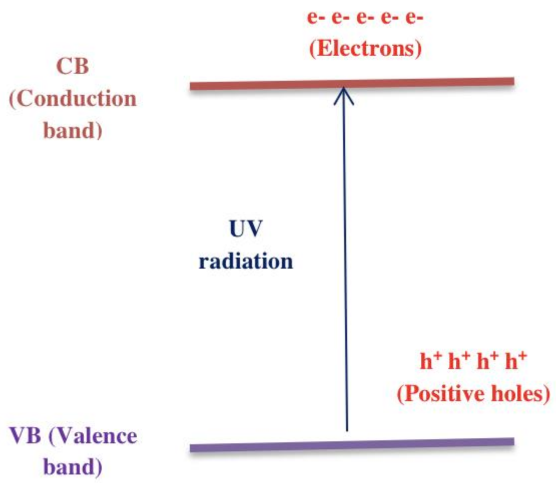


Figure 2. Shifting of electrons from valence to conductance band [14].

The photocatalytic process is initiated by the irradiation of a semiconductor with light with sufficient energy to equal or achieve the band gap, causing the excitation of an electron from the valence band to the conduction band [56,57]. Efficient visible light active photocatalysts consist of several characteristics including the ability of dopants to absorb visible light through CB control of semiconductors, either by establishing an isolated state below the CB or by narrowing the band gap by positively moving the CB bottom; the VB of semiconductors must be in a deeper position upon band engineering, and the photocatalyst surface must be modified by co-catalysts to achieve efficient oxygen reduction [58].

The photocatalyst degradation as shown in Figure 3 occurs under the influence of visible light. Photocatalytic materials will generate photo-oxidation-reduction processes, which entails oxidation from photogenerated holes (h^+) in the valance band (VB) and reduction from photogenerated electrons (e^-) in the conduction band (CB) on the surface. The chemical reaction dissolves the oxygen and water molecules absorbed on the surface. This interaction when energized at higher energy states with light will decompose and oxidize the organic substance, and inorganic compounds such as CO_2 and H_2O [59] will improve the air quality in the environment used in the building and construction industry.

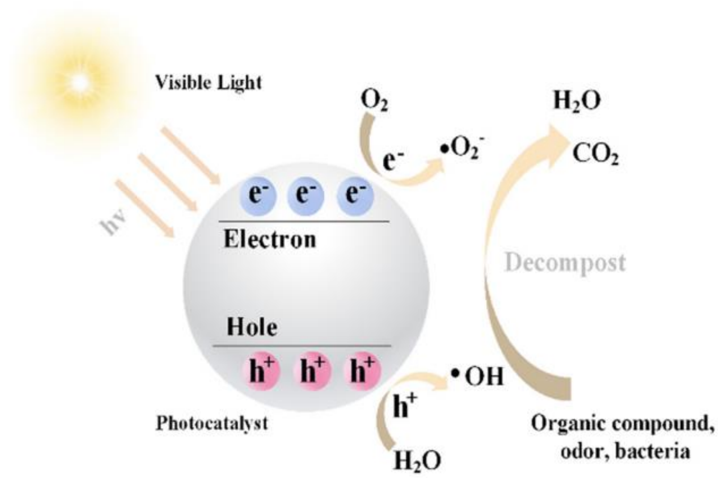


Figure 3. Mechanism of photocatalyst degradation exposed to visible light [59].

Titanium dioxide (TiO_2) is suitable for photocatalysis for a variety of reasons, including its higher photocatalytic efficiency, wide band gap (~3.2 eV), timely availability, low toxicity, low cost, and chemical stability. Various applications of TiO_2 photocatalysis include water treatment, removal of trace metal, water splitting, self-cleaning, antibacterial degradation, self-sterilization, etc. Based on the photocatalyst mechanism processes stated above, the reaction mechanism of TiO_2 comprises Equations (1)–(8) [60].

$$TiO_2 + hv\ (UV) = TiO_2\ (eCB^- + hVB^+) \tag{1}$$

$$TiO_2(hVB^+) + H_2O = TiO_2 + H^+ + OH^\bullet \tag{2}$$

$$TiO_2(hVB^+) + OH^- = TiO_2 + OH^\bullet \tag{3}$$

$$TiO_2(eCB^-) + O_2 = TiO_2 + O_2^{-\bullet} \tag{4}$$

$$O_2^{-\bullet} + H^+ = HO_2^\bullet \tag{5}$$

$$Dye + OH^\bullet = degradation \tag{6}$$

$$Dye + hVB^+ = oxidation \tag{7}$$

$$Dye + eCB^- = reduction \tag{8}$$

Several studies have investigated the photocatalyst reaction in different applications. Carolina Belver et al. [61] reported that numerous inorganic nanomaterials such as metal and metal oxide materials, carbon compounds, zeolite, filtration, and membranes have high reactivity, high functionalization, large specific surface area, and size-dependent properties that are suitable for application in water purification. This technology relies on the availability of a semiconductor that can be stimulated by light with a higher energy than its band gap, resulting in the production of energy-rich electron–hole pairs capable of engaging in redox activities. Anchal Rana et al. [62] reported that metal oxides have been extensively utilized in wastewater treatment; however, because of restrictions, various adjustments are required to make them efficient photocatalysts. Zhu et al. [63] discussed the removal of heavy metal pollutants through the excellent absorption of Zn^{2+} and Fe^{3+}. Several studies investigate the photocatalyst reaction on cement and concrete surfaces. Saffirah et al. [64] reported that the addition of TiO_2 nanoparticles as a photocatalyst for geopolymer paste improve its self-cleaning ability on a concrete surface. This study demonstrated that the self-cleaning characteristics of geopolymers are enhanced by the incorporation of nanoparticles of photocatalytic material [65] because nano-TiO_2 functions as a photocatalyst on the surface of cement concrete. Yang et al. discussed that the maximum photodegradation with the presence of geopolymer boosted the potential of degradation [66].

Extensive study has been conducted in recent years on the effective removal of harmful chemicals and pathogenic bacteria from wastewater using semiconductor photocatalysis, and because electrons and holes in semiconductor materials are activated by light, photocatalytic technology is used to degrade organic wastes or decrease CO_2 for tiny molecule organics. There has been, however, less research using the combination of geopolymer process and photocatalyst as coating materials. Photocatalytic coating materials have the ability to reduce air pollution by converting nitrogen oxide (NO_x) concentrations to less dangerous levels over time while keeping their visual appeal. Table 2 summarizes the applications of photocatalyst technology from previous research.

3.2. Photocatalyst Precursor Materials

Nanomaterials are particles that have one or more exterior dimensions between 1 to 100 nm. Nanomaterials are classified based on the number of dimensions of a material that fall beyond the nanoscale (100 nm) range. Nanoparticles are particles smaller than 100 nm. Nanoparticles have novel or improved size-dependent characteristics as compared with large particles of the same substance. Based on their origin, nanomaterial sources may be divided into three major categories: (i) nanomaterials created as a consequence of industrial activities, such as nanoparticles from car engine exhaust, welding gases, combustion processes, and certain natural processes, such as forest fires; (ii) nanomaterials that have been produced by humans to possess specific characteristics for certain uses; and (iii) nanomaterials created by nature and present in the bodies of creatures and inseminated nanomaterials [72].

Table 2. Summary of photocatalyst technology applications.

Author	Photocatalyst Precursor	Application	Finding
Jdm et al. 2013 [67]	TiO_2 anatase Titanyl sulfate	▪ removal of toxic organic ▪ removal organic contaminants from wastewater	Titanyl sulphate results in high photocatalyst activity.
Maniasaran et al. 2020 [68]	TiO_2	▪ reduces pollutants and acts as self-cleaning surface ▪ air cleaning ▪ water disinfection ▪ self-sterilizing	Geopolymer concrete add TiO_2 is superior in self-cleaning.
M. Mondragon-Figueroa et al. 2019 [69]	TiO_2	▪ Bacteriostatic removal	Metakaolin geopolymer add TiO_2 improved antimicrobial testing.
Isabel Bravo et al. 2019 [70]	Titania	▪ Degradation of dyes in wastewater	Metakaolin and rice husk geopolymer deposited titania degrade 90% of dye pollutants in wastewater.
Anandan et al. 2010 [58]	Zinc oxide	▪ decomposing gaseous acetaldehyde for indoor application.	Efficient ZnO based visible-light photocatalysts, consisting of band-engineering by formation of a solid solution and surface modification of co-catalysts.
Shayegan et al. 2018 [71]	Titanium Dioxide	▪ removal of volatile organic compounds in gas phase	Photocatalyst can eliminate indoor air contaminants effectively at room temperature and contaminants to carbon dioxide and water.

Nanoparticles are interesting because they have unique features such as chemical reactivity and optical behaviour. Nanomaterials are novel materials that have evolved and are being used to improve mechanical strength and improve the characteristics of cementitious materials. In recent years, in order to utilise nanotechnology in construction and building materials, substantial study has been undertaken on the impacts of adding nanoparticles to cementitious materials' characteristics [73]. Photocatalyst nanoparticles are used in building materials, particularly cement, concrete, mortar, paints, and pavement, to increase self-cleaning qualities and cement performance. Nanoparticles were chosen because of their features on cementitious materials, which include great reactivity, ultrafine particle size, and unique physical and chemical properties [74]. The addition of nanosized materials to traditional structural materials is feasible not only for improving the fundamental qualities of cementitious materials but also for adding functionality such as self-cleaning, antibacterial, and pollution-reducing capabilities [75]. Nanoparticles may also be organised in layers on surfaces, resulting in greater surface area and hence increased activity, which is important for a variety of possible applications such as catalysts [72]. Nanoparticles added to cement can be used as an additive or to replace a portion of the cement, providing a substantial contribution to construction materials [49]. Titanium dioxide and zinc oxide are photocatalyst materials that are transparent at the nanoscale but can absorb and reflect UV radiation. Nanoparticles can also be organised in layers on surfaces, resulting in a greater surface area and hence increased activity, which is important for a variety of possible applications such as catalysts [76].

Nanomaterials are essential to photocatalysis research because their characteristics can be precisely tuned, allowing for highly specialised photocatalysis. Nanomaterial photocatalysts are used widely in water splitting to provide clean fuel resources, reduce CO_2 in the environment, remove environmental pollutants, purify air, and self-decontaminate surfaces [67]. By preventing the recombination of charge carriers, the production of heterojunction nanocomposites can modify photocatalysis for applications employing visible light. Thus, nanoparticles are a sort of photocatalyst that merits exploration.

Commonly utilised nanoparticles in cement and concrete products for improving their properties are TiO_2, ZnO, SiO_2, Al_2O_3, and ZrO_2 [77,78]. Photocatalytic capabilities have also been reported for TiO_2 nanoparticles and zinc oxide that have excellent photocatalytic properties [77] that contribute towards environmental pollution.

A novel finding by numerous researchers has regarded the combining of waste by-product materials (e.g., fly ash, rice husk, kaolin and ground granulated blast slag) with nanoparticles, which has demonstrated enhanced mechanical qualities. However, research is still ongoing, and surface coating and surface deterioration prevention have not been fully discussed. Each photocatalyst precursor material routinely employed by researchers is described in the next section.

3.2.1. Titanium Dioxide Nanoparticles Precursor Material

Titanium dioxide (TiO_2) is a white, non-flammable, non-hazardous, thermally stable, and weakly soluble organic solid [79]. TiO_2 is a low-cost, non-hazardous substance that has attracted considerable interest from the coatings industry. TiO_2 is recognized as an excellent photocatalyst for water, a self-cleaning surface material, and an antibacterial substance with hydrophilic characteristics [80]. The three phases of TiO_2 are anatase (tetragonal), rutile (tetragonal), and brookite (orthorhombic) [81]. TiO_2 is a well-known semiconductor with a wide band gap of 3.2 eV for anatase and 3.0 eV for rutile phase [82], but the brookite phase is difficult to acquire [83]. Despite the fact that anatase and rutile have comparable band gaps, anatase has been shown to degrade organic compounds or bio-resistant compounds at a faster rate than rutile [84]. As a result, nanostructured anatase TiO_2 is frequently utilised as a catalyst in photodegradation applications. The excitation of TiO_2 with UV light having an energy higher than the band gap (>3.2 eV) induces the transition of electrons from the valence band to the conduction band and the production of electron/hole pairs [85]. TiO_2 is insoluble in dispersed coatings; hence, performance parameters such as chemical, photochemical, and physical characteristics are mostly governed by the particle size and chemical composition of the pigment's surface [79]. TiO_2 is also remarkable in that it combines a high refractive index with a high degree of visible spectrum transparency [86]. TiO_2 has a much higher refractive index, which means it can scatter light more effectively. When TiO_2 is subjected to an activation light, it can undergo both oxidative and reductive reactions [87]. One of the most demanding criteria for an efficient photocatalyst is that neither photogenerated electrons nor holes be rapidly recombined. By integrating TiO_2 into a TiO_2-based composite such as fly ash or kaolin that inhibits charge recombination, the photocatalytic effectiveness of TiO_2 can be increased. TiO_2 can be employed as a photocatalyst in paints and coatings to minimise environmental pollutants, according to Loh et al. [88]. TiO_2 with nanostructures can be manufactured in a variety of methods depending on the desired material qualities for a given application [89]. For instance, the nano-structured TiO_2 material may be used for self-cleaning and antimicrobial purposes. Hydroxyl radicals may also destroy absorbed organic molecules, including oil on the surface of TiO_2 when exposed to sunlight. As a result, researchers report that most of the photocatalytic process generated by UV light use TiO_2 to assist in the self-cleaning photocatalysis of surfaces [4,13,49].

According to Zulkifly et al. [90], metakaolin geopolymer used as a control specimen exhibits a homogeneous microstructure with few plated-shaped voids; however, metakaolin combined with TiO_2 has a much more compact microstructure. Loh et al. [88] reported that in the absence of light, TiO_2 particle suspensions demonstrated enhanced antifungal activity, as evidenced by the methylene blue decolorization test and the in vitro accelerated biodeterioration test. The antifungal activity of nanoparticle suspensions was evaluated using microbiological techniques. Honarmand et al. [83] showed that TiO_2 nanoparticles (without surfactant) lacked homogeneity and were perhaps amorphous. Titanium alkoxides readily react with water in the absence of surfactants, resulting in amorphous precipitates of TiO_2 nanoparticles. Ratan and Saini [91] investigated the use of nanosized-TiO_2 in the creation of self-cleaning cement concrete and discovered that TiO_2 has the disadvantage that nano-TiO_2 tends to agglomerate in the cement concrete material. However, these limitations should not be used to justify substituting nanosized TiO_2 with microsized TiO_2, as the fundamental disadvantage of TiO_2 at micro sizes is the loss of photoactivity in self-cleaning cement concrete. Geopolymers containing TiO_2 microparticles have a greater absorption capacity, even when the surface areas are small, compared to geopolymers containing no additive. This behaviour is explained by the fact that the surface hydroxyl groups enhance interactions of the TiO_2 micro-particles with H_2O, which in turn presents a significant attraction effect of cationic dyes as reported by M. Mondragon-Figueroa et al. [69]. Numerous studies have found that nano-TiO_2 works as a catalyst in cement hydration processes, resulting in a considerable increase in the rate of early-age hydration of cementitious materials and a change in microstructure that influences the physical and mechanical characteristics of cement-based materials. Meng et al. [92] examined the influence of TiO_2 on the mechanical characteristics of cement mortar. Due to the reduction and modification of the orientation index, the cement mortar's strength enhanced at early ages when nano-TiO_2 was substituted for cement.

Based on the chemical stability of TiO_2, easy availability, high photocatalytic activity, low cost, high dielectric constant, weather ability, anti-microbial properties, and UV radiation stability, the incorporation of nano-TiO_2 as photocatalyst into cement-based material has attracted great exposure and extensive applications.

3.2.2. Zinc Oxide Nanoparticles Precursor Material

Zinc oxide (ZnO) is a white hexagonal crystals, or more commonly a white powder, that possess minimal toxicity and biodegradability. Due to their versatility for use in a variety of applications, ZnO nanoparticles have been the subject of extensive research. ZnO nanoparticles are utilised in a wide range of products and materials, such as glass, plastics, paints, and ceramics. ZnO nanostructures are notable photocatalyst candidates for use in photodegradation because they are inexpensive, nontoxic, and effective at absorbing a substantial proportion of the solar spectrum [17]. Nanomaterials based on ZnO are also used in a variety of developing applications, including electronics and photocatalysis, due to high stability, superior mechanical strength, and high bulk electron mobility [93]. ZnO has been selected as a photocatalyst due to its strong photocatalytic activity and ability to degrade organic molecules by decomposing the organic pollutants. ZnO has been found as an alternative to titania among other nanoparticles studied owing to its photoluminescence capabilities and unique photocatalytic material [4]. The outstanding chemical, electrical, and thermal stabilities of ZnO nanoparticles are attributed to its energy band of 3.37 eV [94]. The addition of metal and metal oxide nanoparticles to the structure of ZnO nanoparticles modify the gap between the conduction band and the valance band, hence possibly enhancing the photocatalytic properties of the material.

According to Nishan and Claire [95], the addition of ZnO into geopolymer paste will enhance the rate of reaction of sodium hydroxide-activated metakaolin and slow the rate of reaction. The findings also reveal the potential of slag and metakaolin-based alkali-activated materials to efficiently encapsulate zinc inside the binder gel, which is essential to the stabilization of waste. Nochaiya et al. [96] observed that increasing the amount of ZnO as an additive material (1 wt.%, 2 wt.% and 5 wt.%) enhanced the compressive strength and physical qualities of concrete at 28 days. According to Zidi et al. [38], adding ZnO to the geopolymer matrix improves its homogeneity. The geopolymer matrix and ZnO have significant interfacial adhesion, resulting in a more compact and dense network. This improvement may be due to the fact that unfilled position have been filled. The phase analysis of zinc oxide doped metakaolin geopolymer paste shows that a glassy process is produced. When 0.3% and 0.5% zinc oxide filler are added to geopolymer samples, the angle shifts slightly to a higher angle. The filler disturbs the geopolymer's side-chain organisation and enriches the amorphous phase. Fariza et al. [95] concluded that ZnO is a versatile material with several important uses, including UV absorption, high photostability, and biodegradability. Additionally, ZnO may be generated in a variety of particle shapes, which impacts its use in advanced techniques and possible applications in a broad range of technical sectors.

Nevertheless, there have been few findings on the integration of ZnO into geopolymer materials as coatings. As a result, research into the influence of zinc oxide nanoparticles on coating application should be pursued. There are limited data on the characterisation of ZnO nanoparticles in comparison with TiO_2 and their impacts on physical-chemical characteristics in geopolymer structures. The development of these types of ZnO and TiO_2 with geopolymers will contribute to the improvement of photocatalytic performance.

4. Coating

4.1. Conventional Coating

Coating materials are used on surfaces such as steel and ceramic to offer protection, corrosion resistance, wear and erosion resistance, a thermal barrier, or aesthetic. Coating materials made from a variety of chemicals and materials, or from a combination of chemicals. Coating materials classified as solid, liquid, or gas, metallic or nonmetallic, organic or inorganic. Coating provides additional protection to the surface and helps keep it looking like new with comparably little upkeep and an easy-to-clean surface. Each component of a coating material has unique properties, such as binders, which function as the principal component as an adhesive to the substrate. The most common components use as coating are binder, additive, pigment, and the carrier fluid or solvent [97]. According to Goncharenko et al. [98], concrete structures may be protected against sulfuric acid corrosion by the use of epoxy coatings. The findings indicate that epoxy-coated concrete has a much longer service life in acidic environments. Other studies Xiao et al. [99] discussed the potential of polyphenol/metal coating on concrete surface for preventing chloride corrosion attack.

We fully addressed numerous studies conducted on various surface coatings of concrete for boosting durability and testing methodologies for determining coating efficiency. Although organic coating materials such as epoxy resins, silane, and acrylic are broadly utilized, there are limitations and downsides such as ease of cracking, peeling, and degradation. Organic matrices also have an ability to discharge vapour pressure which causes concrete damage and coating delamination [100]. Epoxy, resins, acrylics, polytheramides, erethane, and chemical organic coating are frequently used in industrial coating, and they are applied to tanks, pipelines, bridges, marine structures, industrial buildings, and other structures to protect against corrosion, abrasion, surface degradation, and environmental conditions. Epoxy and urethane coatings are resistant to a wide variety of chemical and physical impacts, flexible and low shrinkage but suffer when exposed to the environment [101]. On the other hand, epoxy resins coating depends on the curing agents such as acids and amines. Acrylic coating applied on concrete showed the presence of small amount of VOCs [102]. The analysis indicates that weathering had no effect on the permeability of carbon dioxide, and the material's effect on its vapor permeability was insignificant. This category of substances poses a threat to human health. Therefore, while choosing a coating to be applied, one must consider the coating material's qualities and the environment in which the applications will be utilised.

Based on the research, the conventional coating are useful in protective surface of coating but the uses of epoxy, resins and other chemical organic coating causes harmful and hazardous to human and environment. There are still gaps to fill in the literature.

The next section will elaborate on geopolymer inorganic coatings as alternatives to organic coatings. The potential of geopolymer-based material as coatings was encouraged by the abundance of industrial waste that could be utilised as a source of material for geopolymer coatings.

4.2. Geopolymer Coating

Geopolymer coatings have the potential to preserve surfaces, increase structural longevity, and be applied in high temperature exposure conditions because of its high chemical, mechanical, and thermal resistance qualities, geopolymer ceramic coating offers promise in protective coating and fire resistance. Inorganic coating benefits include excellent temperature resistance, high tensile strength, acid resistance, and environmental friendliness [103]. In geopolymer coating paste, inorganic materials have an aluminosilicates composition with good mechanical properties and high stability. As reported by Davidovits [33], extensive research is being conducted to fully exploit the coating material's potential to encapsulate hazardous waste and low cost. In addition, geopolymer coating also improve the material properties such as strength, acid resistance, corrosion resistance, wear resistance, and as well as protect the environment. These materials have the benefit of achieving strength at room temperature or slightly increased temperatures without requiring the presence of crystalline phases gives great possess of geopolymer coating.

Based on previous studies by Jiang et al. [104], mixing of fly ash, ground granulated blast-furnace slag, metakaolin, and ordinary Portland cement with alkaline activator and the addition of superplasticizer enhanced workability and cracking resistance on concrete surface. On the contrary, Mao et al. [105] deposited metakaolin-based geopolymer coatings on metal through brush air deposition. The research study on the effect of coating behaviours towards curing conditions. Curing in a sealed environment resulted in a restricted geopolymerization response as well as corrosion of the uncoated surface in the metal substrates. Coatings cured at 80 °C had a good morphology, but coatings cured at 150 °C had minor microcracks due to shrinkage and residual stresses. Falah et al. identified geopolymer nanocomposites capable of eliminating hazardous pollutants from wastewater and the atmosphere [9]. Aguirre-Guerrero et al. [106] summarized that alkaline-activated FA/MK geopolymer mortar was prepared as an interfacial agent to prevent corrosion of reinforcing bars implanted in reduced-scale concrete components. The results led to the relation that MK-based geopolymer coating reduced the corrosion rate the most effectively. Sarumathi et al. [107] reviewed several coatings applied to the surface of concrete, as well as the standard tests used to evaluate the effectiveness of surface coatings. Rosales et al. [108] investigated the photocatalytic activity of a SiO_2@TiO_2 coating and showed the removal of Rhodamine B (RhB), establishing it as a photocatalytic material. Guzman-Aponte et al. [109] demonstrated that the inclusion of up to 10 wt.% titanium dioxide particles did not influence the development of the K-A-S-H gel while the setting time of the material and flowability driven by the inclusion of particles of titanium oxide. Alouani et al. [110] studied the ability of geopolymer powder produced from metakaolin and alkaline activators to react as an adsorbent to remove methylene blue. Loh et al. [88] evaluated suspensions of titanium and zinc nanoparticles for their use in the degradation of pollutants and in protecting concrete structures. The results showed that TiO_2 and ZnO nanoparticles protected calciferous materials from fungal fouling and that light exposure was unnecessary for fungal properties. Table 3 shows a summary of previous research findings using geopolymer for coatings, and the research gap of each study is discussed.

Geopolymer coatings have attracted considerable interest as an alternative protective coating material for surfaces including concrete, steel, and ceramic. Sealing the exposed surface of concrete with coatings has been widely investigated and used to extend the life of civil constructions. By using geopolymer-based materials instead of traditional cement-based ones in the construction and civil engineering sectors, greenhouse gas emissions can be reduced [111]. This geopolymer coating is also an innovative material for surface protection and a competitive replacement for conventional synthetic polymer coatings in building applications. Next chapter will discuss the factor affecting the geopolymer coating. The emergence of geopolymer coatings with photocatalytic properties provides an additional benefit for pollutant reduction, corrosion resistance and self-cleaning properties to protect surface deterioration of the coating.

Table 3. Summary of previous research findings in geopolymer coating.

Authors	Field of Study	Finding Descriptions	Research Gap
Jiang et al. 2022 [104]	Fly ash combined with ground granulated blast-furnace slag, metakaolin and ordinary Portland cement added superplasticizer for geopolymer coating concrete.	The adhesive strength of the recommended GPC mixes varied from 1.5 to 3.4 MPa and fully met the surface protection criteria.	The study shows the effect of geopolymer coating for surface protection in building construction and several gaps can be filled regarding the function of superplasticizer. Their properties for enhancing adhesion strength are not fully discussed.
Mao et al. 2020 [105]	Metakaolin-based geopolymer coatings on metal by air brush deposition.	Applied metakaolin-based geopolymer spraying on hot aluminium and steel metal surfaces (40–150 °C) at sealed and unsealed conditions for thermal protection applications.	The study focuses on the effects of curing conditions but gives less explanation regarding the effect of curing temperature on the surface deterioration of metal substrates.
Rosales et al. 2020 [108]	Development of a $SiO_2@TiO_2$ coating applicable to cement-based materials for hydrophobic applications.	The photocatalytic activity of the $SiO_2@TiO_2$ coating showed a removal of RhB establishing itself as a photocatalytic material.	Hydrophobic effect relevant for self-cleaning application on coating but this research focus on conventional coating with different synthesis method such as sol gel and hydrothermal. No research study on the effect of hydrophobic towards geopolymer coating.
Falah et al. 2020 [9]	The effective activation and utilization of metakaolin as an alkali activated geopolymer precursor and its use surface protection.	Geopolymer nanocomposites capable of eliminating hazardous pollutants from wastewater or the atmosphere.	This review of the utilization of metakaolin geopolymer for surface protection did not cover the strength of coating in term of adhesion, corrosion resistance, or abrasion.
Alouani et al. 2019 [110]	The ability of geopolymer powder produced from metakaolin and alkaline activators to react as an adsorbent to remove methylene blue.	Geopolymer has high selectivity and considered an economical adsorbent for the elimination of methylene blue.	The adsorption of MB in geopolymer explained by pseudo-second-order kinetic model, but there is less literature on the testing to eliminate dyes.
Loh et al. 2018 [88]	Titanium and zinc nanoparticle suspensions use in degradation of pollutants and protection of built concrete structures.	TiO_2 and ZnO nanoparticles protected calciferous materials from fungal fouling and light exposure was not necessary for antifungal activity.	The focusing method are incorporated nanoparticles into OPC for concrete building. No relevant information on strength and surface characteristics after addition of nanoparticles.
Aguirre-Guerrero et al. 2017 [106]	Geopolymer mortars containing fly ash and metakaolin as coatings for reinforced concrete against chloride-induced corrosion.	The findings led to the conclusion that the MK based geopolymer coating performed the best, reducing the corrosion rate compared to concrete without coating.	The corrosion reinforcing steel exposed only to the chloride environment and less investigates on the surface deterioration after concrete exposed to chloride attack.
Guzman-Aponte et al. 2017 [109]	TiO_2 addition into the physical and mechanical characteristics of a geopolymer process derived on metakaolin.	Based on the findings, the addition of TiO_2 particles at up to 10 wt.% had no effect on the development of the KASH gel.	The research mentions the physical and mechanical characterization but says less about the strength behaviour of coating after the addition of TiO_2 into the geopolymer.

5. Factor Affecting Geopolymer Paste

Geopolymerization reaction required numerous parameters to consider. One of the most significant variables in this regard is determining the physical properties and chemical composition of the raw material, since this influences the activator's alkaline degree.

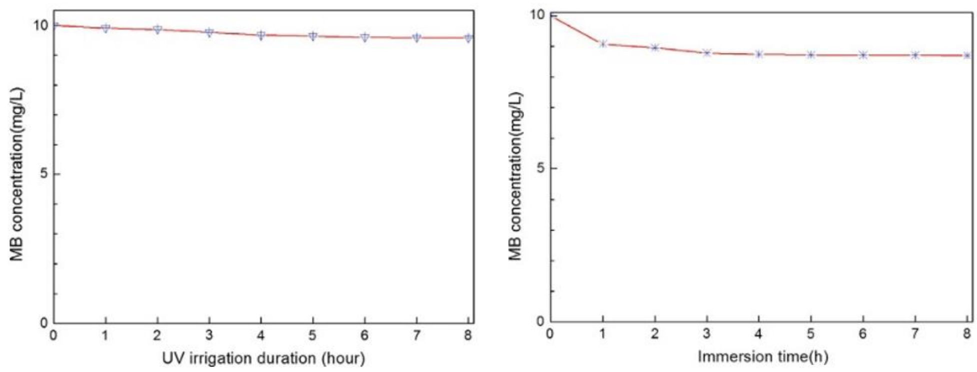
Though since raw materials differ from batch to batch (minerals or waste materials, for example), it is critical to thoroughly analyse the samples before adjusting the composition and amount of the activating solution based on solid to liquid ratio, and percentage of photocatalyst precursor. The solid-to-liquid (S/L) ratio influences the characteristics of geopolymer paste. Jaya et al. [112] exposed the influence of numerous S/L ratio ranging from 0.6 to 0.8. It was found that 0.76 is the optimal ratio for S/L ratios. Compared to the others, the surface of geopolymer with an optimum S/L ratio was more homogenous, dense, and porous due to high strength. The high compressive strength proved by the phase analysis revealed no traces of zeolite crystal peaks. Guzman-Aponte et al. [113] on the other hand, employed a 0.8 ratio to achieve good workability in their investigation. Previous research from Guzman-Aponte et al. [109] discussed liquid/solid ratio (L/S) was also varied in three levels, 0.35 (dry consistency), 0.40 (mean consistency), and 0.45 (fluid consistency). The results indicate that increased liquid content can promote the speed of dissolution of the Al and Si species of the precursor, but it obstructs the processes of polycondensation. Albidah et al. [47] study the solid to liquid ratio ranging from 0.3 to 0.8 shows hen alkaline solids to MK ratio increased from 0.37 to 0.41, the compressive strength dropped from 58.5 to 39.7 MPa. The strength reduction can be attributed to the excess amount of silica content as the SiO_2/Al_2O_3 molar ratio was increased from 2.69 to 2.75, which can contribute to the impedance of geopolymerization process. Yaacob et al. [114] used a S/L ratio of 0.33 to produce a geopolymer coating with maximum adhesion strength of 3.8 MPa.

In addition to the solid-to-liquid ratio, the percentage of photocatalyst precursor also plays an important role in enhancing the photocatalytic performance of geopolymer pastes suitable for coating. Wang et al. [115] discovered that 5 wt.% TiO_2 resulted in the most efficient photocatalytic based on methylene blue colour alteration. Setting time and leaching studies demonstrate that Zn has a much stronger retarding impact on reaction kinetics in Na-activated geopolymers compared with K-activated geopolymers. However, Na-activated geopolymers have a better fixing ability to Zn. In the Zn-substituted geopolymer system, Al_2O_3/M_2O ratio was 0.8 and ZnO/M_2O ratio was 0.2 respectively. According to Guzmán-Aponte et al. [109] the percentage of TiO_2 addition as a function of cement was revised at three levels; 0 wt.%, 5 wt.% and 10 wt.%. This amount of TiO_2 effect the performance of photocatalytic efficiency by improving the carbonation process and band gap energy. The results show that a percentage of TiO_2 up to 10% has no influence on the mechanical characteristics of geopolymer and the production of the K-A-S-H gel. According to Zidi et al. [38] the optimal quantity of nano-ZnO to enhance the mechanical and structural of metakaolin geopolymer was 0.5 wt.%. The incorporation of nano-ZnO enhanced pulsed velocity and boosted compressive strength from 30 to 38 MPa. The optimum percentage of ZnO was found to be 0.5 wt.% according to research from Wang et al. [115], ZnO responded fully and formed amorphous products after 7 days of curing. The crystallinity phases of a metakaolin geopolymer paste are unaffected by the addition of ZnO nanoparticles as a photocatalyst. Aside from that, Zailan et al. [64] prepared the TiO_2 geopolymer paste by varying the percentage of nano-TiO_2, which are 5.0 wt.%, 10.0 wt.% and 15.0 wt.%. The study discovered the methylene blue discoloration after exposure to sunlight up to 150 min with great photocatalytic effect. Strini et al. [116] added 3% of TiO_2 by weight paste into the fly ash and metakaolin geopolymer. The findings demonstrated that geopolymer binders can be effective catalyst support matrices for the coating applications. Based on the studies from other researchers, less literature review regarding addition of photocatalyst precursor into the geopolymer paste for coating characterization and this gap can be fulfilled.

6. Photocatalyst Degradation Evaluation

The photodegradation approach causes difficulties in terms of eliminating products created in the water environment during the photocatalytic degradation process. Several research initiatives have concentrated on the elements of materials produced by distributing photocatalysts in native or synthetic materials and generating photocatalytic matrices.

Saufi et al. [117] monitoring the decomposition efficiency of chemical molecules in discoloration of methylene blue using the perlite-based geopolymer. The results demonstrate that perlite-based geopolymer with UV light has improved adsorption and photodegradation properties for methylene blue molecules compared to perlite-based geopolymer without UV radiation. This efficacy is attributed to a catalytic activity rather than a simple linear combination of perlite-based geopolymer powder with UV irradiation for methylene blue removal from aqueous solution [117]. Loh et al. [88] results demonstrated that even in the absence of light, both ZnO and TiO_2 particle suspensions displayed antimicrobial behaviour in the MB decolorization and fast biodeterioration tests. ZnO exceeded TiO_2 in photocatalytic antifungal activity. Jdm et al. [67] carried out experiment in aqueous solution whereas methylene blue was degraded by TiO_2 anatase, TiO_2 from titanyl sulphate, geopolymer, geopolymer with anatase, and geopolymer with titanyl sulphate. The geopolymer mixture containing titanyl sulphate had the highest photocatalyst activity. Throughout the 60-min duration of the reaction, the MB component's solution absorption intensity decreased. This occurrence revealed that in the presence of another substance, MB molecules were invaded and driven out. Without the presence of geopolymer, the interaction of MB with the materials is very slow, which might result in poor MB component conversion. As a result, UV irradiation of MB ink on materials without geopolymer and with anatase produces an exceptionally white colour, in contrast to other samples that were colourless in this experiment [67]. Kaya et al. [118] explored the effectiveness of geopolymers based on metakaolin and red mud, testing the adsorption and photocatalytic decomposition of methylene blue to demonstrate their self-cleaning capabilities. The photocatalytic activity increased as anatase concentration increased. Geopolymer with a 3.7 wt.% anatase loading exhibited a greater photocatalytic degradation constant than pure anatase. Strini et al. [116] studied photocatalytic compounds utilising cementitious binders as catalytic supports networks, with geopolymer matrices obtained from fly ash and metakaolin. The photocatalytic activity was evaluated based on the degree to which NO_x was degraded in the air, and the results were evaluated in relation to a reference of Portland cement. Depending on the aluminosilicate source, the samples exhibit significantly diverse photocatalytic activity, and the results showed that the photocatalytic activity of fly ash-based geopolymer was the most suitable for curing at room temperature, whereas geopolymer based on metakaolin also shown promising photocatalytic activity [116]. Zhang et al. [119] also focused on a geopolymer derived from fly ash as raw material and employed it as a novel photocatalyst for wastewater treatment application via methylene blue decomposition. Under UV light, the fly ash-based geopolymer catalyst had a degradation efficiency of MB dye at 93%. This was feasible due to the general synergistic effect of adsorption and semiconductor photocatalysis [119]. The results supported by studies from Isabel Bravo et al. [70] identify titania- after 10 h of exposure coated geopolymer spheres showed dye photodegradation activity of 90%, validating the products as sustainable wastewater treatment materials. Chen et al. [120] conduct an analysis of the performance of the deterioration in terms of the total degradation of the MB over time. Figure 4 displays the methylene blue (MB) colour change under UV light with and without photocatalyst precursor [120]. The inclusion of photocatalyst precursors (TiO_2) deteriorated the MB solution. This revealed that TiO_2 is highly useful for the geopolymer coating in this investigation.



(**a**) (**b**)

Figure 4. The methylene blue (MB) colour change under UV light (**a**) without photocatalyst precursor; (**b**) with photocatalyst precursor [120].

Guzman et al. [113] demonstrates the concentration of methylene blue (MB) under dark and UV-irradiated settings for metakaolin geopolymer paste supplemented by zinc oxide (ZnO). Throughout the first 30 min of photoactivation, the residual concentration of ZnO nanostructures changed as shown in Figure 5 [113].

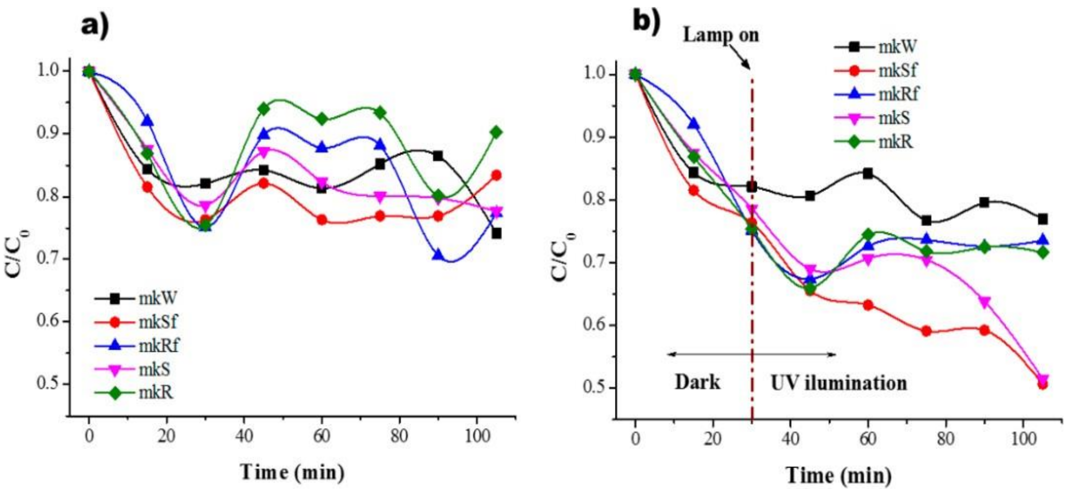


Figure 5. Methylene blue (MB) degradation (**a**) in dark conditions and (**b**) under UV light for the metakaolin paste with zinc oxide (ZnO) nanostructure [113].

Photocatalytic activity is widely known to occur at the surface of a photocatalyst. As a result, the surface area of coating, which is affected by photocatalyst precursor nanoparticle size, film shape, and thickness, has an influence on photocatalytic reactivity. The results showed that the ZnO-TiO_2 nano-composite catalyst exhibits ZnO or TiO_2 nanoparticles in terms of photocatalytic capabilities. The enhanced photocatalytic activities were related to the prevention of electron-hole pair recombination and synergistic effect between hexagonal ZnO and TiO_2 phases [93].

7. Conclusions

This review thoroughly discussed various studies carried out regarding the geopolymer with photocatalyst effect to overcome problem regarding organic dyes and alternative environmentally friendly construction material such as geopolymer concrete has been developed. There are still gaps to fill in the literature as researchers are more focusing on the physical and mechanical properties and have not been concentrated on the formulations of coatings that are durable and extend the lives of surfaces against deterioration. It has also not been explored how well the cleanliness of building surfaces can be maintained, and the air pollution also can be reduced using geopolymer photocatalyst coatings. Based on the previous research, photocatalyst materials have significance for additions to geopolymer pastes. However, the usage of geopolymers for self-cleaning applications and coatings has not been fully investigated, for instance with adhesion tests, abrasion tests, fire resistance limit testing, and UV-vis analysis. It is conceivable to identify the effects of photocatalytic precursors on these properties. This brief analysis proves that there is still much opportunity for study and improvement in the realm of zinc oxide and titanium dioxide as additions for geopolymer coating applications.

Author Contributions: Conceptualization, L.J., R.A.R. and M.M.A.B.A.; data curation, L.J., R.A.R., P.V. and A.B.; formal analysis, L.J., R.A.R. and S.Z.A.R.; investigation, L.J., P.V., A.V.S., A.B. and S.Z.A.R.; methodology, L.J., R.A.R. and M.M.A.B.A.; project administration, R.A.R., T.I. and H.C.Y.; validation, R.A.R., T.I. and M.M.A.B.A.; writing of review and editing, L.J., R.A.R., M.M.A.B.A., A.V.S. and H.C.Y. All authors have read and agreed to the published version of the manuscript.

Funding: This study was supported by the Center of Excellence Geopolymer and Green Technology (CEGeoGTech) UniMAP and Faculty of Mechanical Engineering and Technology, UniMAP. This research was also supported by TUIASI from the University Scientific Research Fund (FCSU).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors wish to extend their gratitude to the Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis (UniMAP), for their support with providing all the necessary facilities.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Pillay, D.L.; Olalusi, O.B.; Awoyera, P.O.; Rondon, C.; Echeverría, A.M.; Kolawole, J.T. A Review of the Engineering Properties of Metakaolin Based Concrete: Towards Combatting Chloride Attack in Coastal/Marine Structures. *Adv. Civ. Eng.* **2020**, *2020*, 1–13. [CrossRef]
2. Petroche, D.M.; Ramirez, A.D. The Environmental Profile of Clinker, Cement, and Concrete: A Life Cycle Perspective Study Based on Ecuadorian Data. *Buildings* **2022**, *12*, 311. [CrossRef]
3. Zhang, J.; Tian, B.; Wang, L.; Xing, M.; Lei, J. Mechanism of Photocatalysis. In *Photocatalysis. Lecture Notes in Chemistry*; Springer: Singapore, 2018; Volume 100.
4. Zailan, S.N.; Mahmed, N.; Abdullah, M.M.A.B.; Rahim, S.Z.A.; Halin, D.S.C.; Sandu, A.V.; Vizureanu, P.; Yahya, Z. Potential Applications of Geopolymer Cement-Based Composite as Self-Cleaning Coating: A Review. *Coatings* **2022**, *12*, 133. [CrossRef]
5. Burghardt, T.E.; Pashkevich, A.; Zakowska, L. Influence of Volatile Organic Compounds Emissions from Road Marking Paints on Ground-Level Ozone Formation: Case Study of Kraków, Poland. *Transp. Res. Procedia* **2016**, *14*, 714–723. [CrossRef]
6. Jiménez-López, A.M.; Hincapié-Llanos, G.A. Identification of Factors Affecting the Reduction of VOC Emissions in the Paint Industry: Systematic Literature Review-SLR. *Prog. Org. Coat.* **2022**, *170*, 106945. [CrossRef]
7. Zhu, A. Feasibility Study on Novel Fire-Resistant Coating Materials. Master's Thesis, Missouri University of Science and Technology, Rolla, MO, USA, 2020.
8. Warid Wazien, A.Z.; Mustafa, M.; Abdullah, A.B.; Razak, R.A.; Rozainy, M.M.A.Z.R.; Faheem, M.; Tahir, M.; Faris, M.A.; Hamzah, H.N. Review on Potential of Geopolymer for Concrete Repair and Rehabilitation. *MATEC Web Conf.* **2016**, *78*, 01065. [CrossRef]
9. Falah, M.; Mackenzie, K.J.D. Photocatalytic Nanocomposite Materials Based on Inorganic Polymers (Geopolymers): A Review. *Catalysts* **2020**, *10*, 1158. [CrossRef]
10. Khaki, M.R.D.; Shafeeyan, M.S.; Raman, A.A.A.; Daud, W.M.A.W. Application of Doped Photocatalysts for Organic Pollutant Degradation—A Review. *J. Environ. Manag.* **2017**, *198*, 78–94. [CrossRef]
11. Mashuri, S.I.S.; Ibrahim, M.L.; Kasim, M.F.; Mastuli, M.S.; Rashid, U.; Abdullah, A.H.; Islam, A.; Asikin-Mijan, N.; Tan, Y.H.; Mansir, N.; et al. Photocatalysis for Organic Wastewater Treatment: From the Basis to Current Challenges for Society. *Catalysts* **2020**, *10*, 1260. [CrossRef]
12. Garcia-Segura, S.; Brillas, E. Applied Photoelectrocatalysis on the Degradation of Organic Pollutants in Wastewaters. *J. Photochem. Photobiol. C Photochem. Rev.* **2017**, *31*, 1–35. [CrossRef]
13. Rabajczyk, A.; Zielecka, M.; Klapsa, W.; Dziechciarz, A. Self-Cleaning Coatings and Surfaces of Modern Building Materials for the Removal of Some Air Pollutants. *Materials* **2021**, *14*, 2161. [CrossRef] [PubMed]

14. Joshi, N.C.; Gururani, P.; Gairola, S.P. Metal Oxide Nanoparticles and Their Nanocomposite-Based Materials as Photocatalysts in the Degradation of Dyes. *Biointerface Res. Appl. Chem.* **2022**, *12*, 6557–6579.
15. Dharma, H.N.C.; Jaafar, J.; Widiastuti, N.; Matsuyama, H.; Rajabsadeh, S.; Othman, M.H.D.; Rahman, M.A.; Jafri, N.N.M.; Suhaimin, N.S.; Nasir, A.M.; et al. A Review of Titanium Dioxide (TiO_2)-Based Photocatalyst for Oilfield-Produced Water Treatment. *Membranes* **2022**, *12*, 345. [CrossRef] [PubMed]
16. Anucha, C.B.; Altin, I.; Bacaksiz, E.; Stathopoulos, V.N. Titanium Dioxide (TiO_2)-Based Photocatalyst Materials Activity Enhancement for Contaminants of Emerging Concern (CECs) Degradation: In the Light of Modification Strategies. *Chem. Eng. J. Adv.* **2022**, *10*. [CrossRef]
17. Ong, C.B.; Ng, L.Y.; Mohammad, A.W. A Review of ZnO Nanoparticles as Solar Photocatalysts: Synthesis, Mechanisms and Applications. *Renew. Sustain. Energy Rev.* **2018**, *81*, 536–551. [CrossRef]
18. Samadi, M.; Zirak, M.; Naseri, A.; Khorashadizade, E.; Moshfegh, A.Z. Recent Progress on Doped ZnO Nanostructures for Visible-Light Photocatalysis. *Thin Solid Films.* **2016**, *605*, 2–19. [CrossRef]
19. Lee, K.M.; Lai, C.W.; Ngai, K.S.; Juan, J.C. Recent Developments of Zinc Oxide Based Photocatalyst in Water Treatment Technology: A Review. *Water Res.* **2016**, *88*, 428–448. [CrossRef]
20. Sharifi, T.; Crmaric, D.; Kovacic, M.; Popovic, M.; Rokovic, M.K.; Kusic, H.; Jozić, D.; Ambrožić, G.; Kralj, D.; Kontrec, J.; et al. Tailored $BiVO_4$ for Enhanced Visible-Light Photocatalytic Performance. *J. Environ. Chem. Eng.* **2021**, *9*, 106025. [CrossRef]
21. Shan, L.; Lu, C.; Dong, L.; Suriyaprakash, J. Efficient Facet Regulation of $BiVO_4$ and Its Photocatalytic Motivation. *J. Alloys Compd.* **2019**, *804*, 385–391. [CrossRef]
22. Shafiq, I.; Hussain, M.; Shafique, S.; Rashid, R.; Akhter, P.; Ahmed, A.; Jeon, J.-K.; Park, Y.-K. Oxidative Desulfurization of Refinery Diesel Pool Fractions Using $LaVO_4$ Photocatalyst. *J. Ind. Eng. Chem.* **2021**, *98*, 283–288. [CrossRef]
23. Zhang, M.; Xu, J.; Chen, M. Novel Z-Scheme $LaVO_4/Bi_3O_4Cl$ Heterojunctions for Highly Efficient Degradation of Ofloxacin under Visible Light Irradiation. *J. Alloys Compd.* **2022**, *925*, 166653. [CrossRef]
24. Qi, C.; Bao, W.; Wang, L.; Li, H.; Wu, W. Study of the V_2O_5-WO_3/TiO_2 Catalyst Synthesized from Waste Catalyst on Selective Catalytic Reduction of NOx by NH_3. *Catalysts* **2017**, *7*, 110. [CrossRef]
25. Burduhos Nergis, D.D.; Vizureanu, P.; Ardelean, I.; Sandu, A.V.; Corbu, O.C.; Matei, E. Revealing the Influence of Microparticles on Geopolymers' Synthesis and Porosity. *Materials* **2020**, *13*, 3211. [CrossRef] [PubMed]
26. Meor Ahmad Tajudin, M.A.F.; Abdullah, M.M.A.B.; Sandu, A.V.; Nizar, K.; Moga, L.; Neculai, O.; Muniandy, R. Assessment of Alkali Activated Geopolymer Binders as an Alternative of Portland Cement. *Mater. Plastice.* **2017**, *54*, 145–154.
27. Vizureanu, P.; Samoila, C.; Cotfas, D. Materials Processing using Solar Energy. *Environ. Eng. Manag. J.* **2009**, *8*, 301–306. [CrossRef]
28. Azimi, E.A.; Abdullah, M.M.A.B.; Vizureanu, P.; Salleh, M.A.A.M.; Sandu, A.V.; Chaiprapa, J.; Yoriya, S.; Hussin, K.; Aziz, I.H. Strength Development and Elemental Distribution of Dolomite/Fly Ash Geopolymer Composite under Elevated Temperature. *Materials* **2020**, *13*, 1015. [CrossRef]
29. Burduhos Nergis, D.D.; Vizureanu, P.; Corbu, O. Synthesis and Characteristics of Local Fly Ash Based Geopolymers Mixed with Natural Aggregates. *Rev. De Chim.* **2019**, *70*, 1262–1267. [CrossRef]
30. Duxson, P.; Fernández-Jiménez, A.; Provis, J.L.; Lukey, G.C.; Palomo, A.; van Deventer, J.S.J. Geopolymer Technology: The Current State of the Art. *J. Mater. Sci.* **2007**, *42*, 2917–2933. [CrossRef]
31. Blissett, R.S.; Rowson, N.A. A Review of the Multi-Component Utilisation of Coal Fly Ash. *Fuel* **2012**, *97*, 1–23. [CrossRef]
32. Siyal, A.A.; Shamsuddin, M.R.; Khan, M.I.; Rabat, N.E.; Zulfiqar, M.; Man, Z.; Siame, J.; Azizli, K.A. A Review on Geopolymers as Emerging Materials for the Adsorption of Heavy Metals and Dyes. *J. Environ. Manag.* **2018**, *224*, 327–339. [CrossRef]
33. Davidovits, J. Geopolymers. *J. Therm. Anal.* **1991**, *37*, 1633–1656. [CrossRef]
34. Saloma; Hanafiah; Elysandi, D.O.; Meykan, D.G. Effect of $Na_2SiO_3/NaOH$ on Mechanical Properties and Microstructure of Geopolymer Mortar Using Fly Ash and Rice Husk Ash as Precursor. *AIP Conf. Proc.* **2017**, *1903*, 050013.
35. Al Bakri, A.M.; Kamarudin, H.; Bnhussain, M.; Nizar, I.K.; Mastura, W. Mechanism and Chemical Reaction of Fly Ash Geopolymer Cement—A Review. *J. Asian Sci. Res.* **2011**, *1*, 247–253.
36. Castillo, H.; Collado, H.; Droguett, T.; Vesely, M.; Garrido, P.; Palma, S. State of the Art of Geopolymers: A Review. *E-Polymer* **2022**, *22*, 108–124. [CrossRef]
37. Adewuyi, Y.G. Recent Advances in Fly-Ash-Based Geopolymers: Potential on the Utilization for Sustainable Environmental Remediation. *ACS Omega* **2021**, *6*, 15532–15542. [CrossRef] [PubMed]
38. Zidi, Z.; Ltifi, M.; ben Ayadi, Z.; Mir, L.E.L.; Nóvoa, X.R. Effect of Nano-ZnO on Mechanical and Thermal Properties of Geopolymer. *J. Asian Ceram. Soc.* **2020**, *8*, 1–9. [CrossRef]
39. Cao, R.; Fang, Z.; Jin, M.; Shang, Y. Study on the Activity of Metakaolin Produced by Traditional Rotary Kiln in China. *Minerals* **2022**, *12*, 365. [CrossRef]
40. Rocha, J.; Klinowski, J. *Physics and Chemistry of Minerals, 295i and 27A1 Magic-Angle-Spinning NMR Studies of the Transformation of Kaolinite*; University of Cambridge: Cambridge, UK, 1990; Volume 17.
41. De Rossi, A.; Simão, L.; Ribeiro, M.J.; Novais, R.M.; Labrincha, J.A.; Hotza, D.; Moreira, R.F.P.M. In-Situ Synthesis of Zeolites by Geopolymerization of Biomass Fly Ash and Metakaolin. *Mater. Lett.* **2019**, *236*, 644–648. [CrossRef]
42. Zhuang, X.Y.; Chen, L.; Komarneni, S.; Zhou, C.H.; Tong, D.S.; Yang, H.M.; Yu, W.H.; Wang, H. Fly Ash-Based Geopolymer: Clean Production, Properties and Applications. *J. Clean. Prod.* **2016**, *125*, 253–267. [CrossRef]

43. Kalombe, R.M.; Ojumu, V.T.; Eze, C.P.; Nyale, S.M.; Kevern, J.; Petrik, L.F. Fly Ash-Based Geopolymer Building Materials for Green and Sustainable Development. *Materials* **2020**, *13*, 5699. [CrossRef]
44. Prochon, P.; Zhao, Z.; Courard, L.; Piotrowski, T.; Michel, F.; Garbacz, A. Influence of Activators on Mechanical Properties of Modified Fly Ash Based Geopolymer Mortars. *Materials* **2020**, *13*, 1033. [CrossRef] [PubMed]
45. Humad, A.M.; Kothari, A.; Provis, J.L.; Cwirzen, A. The Effect of Blast Furnace Slag/Fly Ash Ratio on Setting, Strength, and Shrinkage of Alkali-Activated Pastes and Concretes. *Front. Mater.* **2019**, *6*, 9. [CrossRef]
46. Abbas, R.; Khereby, M.A.; Ghorab, H.Y.; Elkhoshkhany, N. Preparation of Geopolymer Concrete Using Egyptian Kaolin Clay and the Study of Its Environmental Effects and Economic Cost. *Clean Technol. Environ. Policy* **2020**, *22*, 669–687. [CrossRef]
47. Albidah, A.; Alghannam, M.; Abbas, H.; Almusallam, T.; Al-Salloum, Y. Characteristics of Metakaolin-Based Geopolymer Concrete for Different Mix Design Parameters. *J. Mater. Res. Technol.* **2021**, *10*, 84–98. [CrossRef]
48. Ionescu, B.A.; Lăzărescu, A.-V.; Hegyi, A. The Possibility of Using Slag for the Production of Geopolymer Materials and Its Influence on Mechanical Performances—A Review. *Proceedings* **2020**, *63*, 30.
49. Vignesh, T.; Sumathi, A.; Saravana, K.; Mohan, R. Study on Self-Cleaning Concrete Using Nano-Liquid TiO_2. *Int. J. Eng. Technol.* **2018**, *7*, 860–863. [CrossRef]
50. Ameta, R.; Solanki, M.S.; Benjamin, S.; Ameta, S.C. Photocatalysis. In *Advanced Oxidation Processes for Wastewater Treatment: Emerging Green Chemical Technology*; Elsevier Inc.: Amsterdam, The Netherlands, 2018; pp. 135–175.
51. Zhang, F.; Wang, X.; Liu, H.; Liu, C.; Wan, Y.; Long, Y.; Cai, Z. Recent Advances and Applications of Semiconductor Photocatalytic Technology. *Appl. Sci.* **2019**, *9*, 2489. [CrossRef]
52. Molinari, R.; Lavorato, C.; Argurio, P. Visible-Light Photocatalysts and Their Perspectives for Building Photocatalytic Membrane Reactors for Various Liquid Phase Chemical Conversions. *Catalysts* **2020**, *10*, 1334. [CrossRef]
53. Guo, Y.; Li, H.; Ma, W.; Shi, W.; Zhu, Y.; Choi, W. Photocatalytic Activity Enhanced via Surface Hybridization. *Carbon Energy.* **2020**, *2*, 308–349. [CrossRef]
54. Makuła, P.; Pacia, M.; Macyk, W. How To Correctly Determine the Band Gap Energy of Modified Semiconductor Photocatalysts Based on UV-Vis Spectra. *J. Phys. Chem. Lett.* **2018**, *9*, 6814–6817. [CrossRef]
55. Regmi, C.; Joshi, B.; Ray, S.K.; Gyawali, G.; Pandey, R.P. Understanding Mechanism of Photocatalytic Microbial Decontamination of Environmental Wastewater. *Front. Chem.* **2018**, *6*, 33. [CrossRef] [PubMed]
56. Malato, S.; Fernández-Ibáñez, P.; Maldonado, M.I.; Blanco, J.; Gernjak, W. Decontamination and Disinfection of Water by Solar Photocatalysis: Recent Overview and Trends. *Catal. Today* **2009**, *147*, 1–59. [CrossRef]
57. Li, J.; Yuan, H.; Zhang, W.; Jin, B.; Feng, Q.; Huang, J.; Jiao, Z. Advances in Z-Scheme Semiconductor Photocatalysts for the Photoelectrochemical Applications: A Review. *Carbon Energy* **2022**, *4*, 294–331. [CrossRef]
58. Anandan, S.; Ohashi, N.; Miyauchi, M. ZnO-Based Visible-Light Photocatalyst: Band-Gap Engineering and Multi-Electron Reduction by Co-Catalyst. *Appl. Catal. B Environ.* **2010**, *100*, 502–509. [CrossRef]
59. Wang, Q.; Zheng, K.; Yu, H.; Zhao, L.; Zhu, X.; Zhang, J. Laboratory Experiment on the Nano-TiO_2 Photocatalytic Degradation Effect of Road Surface Oil Pollution. *Nanotechnol. Revis.* **2020**, *9*, 922–933. [CrossRef]
60. Kumar, A. A Review on the Factors Affecting the Photocatalytic Degradation of Hazardous Materials. *Mater. Sci. Eng. Int. J.* **2017**, *1*, 106–114. [CrossRef]
61. Belver, C.; Bedia, J.; Gómez-Avilés, A.; Peñas-Garzón, M.; Rodriguez, J.J. Semiconductor Photocatalysis for Water Purification. In *Nanoscale Materials in Water Purification*; Elsevier: Amsterdam, The Netherlands, 2019; pp. 581–651.
62. Rana, A.; Sudhaik, A.; Raizada, P.; Khan, A.A.P.; van Le, Q.; Singh, A.; Selvasembian, R.; Nadda, A.; Singh, P. An Overview on Cellulose-Supported Semiconductor Photocatalysts for Water Purification. *Nanotechnol. Environ. Eng.* **2021**, *6*, 40. [CrossRef]
63. Zhu, S.; Chen, Y.; Khan, M.A.; Xu, H.; Wang, F.; Xia, M. In-Depth Study of Heavy Metal Removal by an Etidronic Acid-Functionalized Layered Double Hydroxide. *ACS Appl. Mater. Interfaces* **2022**, *14*, 7450–7463. [CrossRef]
64. Zailan, S.N.; Mahmed, N.; Abdullah, M.M.A.B. Photocatalytic Behaviour of TiO_2-Geopolymer Paste under Sunlight. *IOP Conf. Ser. Mater. Sci. Eng.* **2020**, *957*, 012006. [CrossRef]
65. Assi, L.; Carter, K.; Deaver, E.; Anay, R.; Ziehl, P. Sustainable Concrete: Building a Greener Future. *J. Clean. Prod.* **2018**, *198*, 1641–1651. [CrossRef]
66. Yang, X.; Liu, Y.; Yan, C.; Peng, R.; Wang, H. Geopolymer-TiO_2 Nanocomposites for Photocsatalysis: Synthesis by One-Step Adding Treatment versus Two-Step Acidification Calcination. *Minerals* **2019**, *9*, 658. [CrossRef]
67. Jdm, K.; Yusoff, M.M.; Aqilah, N.S. Degradation of Methylene Blue via Geopolymer Composite Photocatalyst. *Solid State Sci. Technol.* **2013**, *21*, 23–30.
68. Maniarasan, S.K.; Santhosh Kumar, V.; Chandrasekaran, P. Index Terms: Application of Titania in Geopolymer Concrete. *Int. J. Sci. Technol. Res.* **2020**, *9*, 806–813.
69. Mondragón-Figueroa, M.; Guzmán-Carrillo, H.R.; Rico, M.Á.; Reyez-Araiza, J.L.; Pineda-Piñón, J.; López-Naranjo, E.J.; Columba-Palomares, M.C.; López-Romero, J.M.; Gasca-Tirado, J.R. Development of a Construction Material for Indoor and Outdoor, Metakaolinite-Based Geopolymer, with Environmental Properties. *J. Mater. Sci. Eng. A* **2019**, *9*, 131–142. [CrossRef]
70. Isabel Bravo, P.; Shimizu, E.; Alvin Malenab, R.; Anne Tigue, A.; Mae Dela Cerna, K.; Isagani Janairo, J.; Angelo Promentilla, M.; Ethelbhert Yu, D. Nanocrystalline Titania Coated Metakaolin and Rice Hull Ash Based Geopolymer Spheres for Photocatalytic Degradation of Dyes in Wastewater. *Orient. J. Chem.* **2019**, *35*, 167–172. [CrossRef]

71. Shayegan, Z.; Lee, C.S.; Haghighat, F. TiO2 Photocatalyst for Removal of Volatile Organic Compounds in Gas Phase—A Review. *Chem. Eng. J.* **2018**, *334*, 2408–2439. [CrossRef]
72. Jeevanandam, J.; Barhoum, A.; Chan, Y.S.; Dufresne, A.; Danquah, M.K. Review on Nanoparticles and Nanostructured Materials: History, Sources, Toxicity and Regulations. *Beilstein J. Nanotechnol.* **2018**, *9*, 1050–1074. [CrossRef] [PubMed]
73. Li, Z.; Ding, S.; Yu, X.; Han, B.; Ou, J. Multifunctional Cementitious Composites Modified with Nano Titanium Dioxide: A Review. *Compos. Part A: Appl. Sci. Manuf.* **2018**, *111*, 115–137. [CrossRef]
74. Ningthoujam, R.; Singh, Y.D.; Babu, P.J.; Tirkey, A.; Pradhan, S.; Sarma, M. Nanocatalyst in Remediating Environmental Pollutants. *Chem. Phys. Impact* **2022**, 4. [CrossRef]
75. Hamidi, F.; Aslani, F. TiO_2-Based Photocatalytic Cementitious Composites: Materials, Properties, Influential Parameters, and Assessment Techniques. *Nanomaterials* **2019**, *9*, 1444. [CrossRef]
76. Smijs, T.G.; Pavel, S. Titanium Dioxide and Zinc Oxide Nanoparticles in Sunscreens: Focus on Their Safety and Effectiveness. *Nanotechnol. Sci. Appl.* **2011**, *4*, 95–112. [CrossRef] [PubMed]
77. Ketan, M.; Jibhenkar, B.; Vaidya, P.V.D.; Waghmare, M.S.S.; Singh, D.P. Eco-Sustainable Pervious Concrete Using Titanium Dioxide. *Int. J. Sci. Res.Dev.* **2015**, *3*, 391–392.
78. Liu, J.; Li, Q.; Xu, S. Influence of Nanoparticles on Fluidity and Mechanical Properties of Cement Mortar. *Constr. Build. Mater.* **2015**, *101*, 892–901. [CrossRef]
79. Jamaludin, L.; Razak, R.A.; al Bakri Abdullah, M.M.; Kusbiantoro, A.; Yahya, Z.; Abdullah, A.; Sandu, A.V. Geopolymer Coating Paste on Concrete for Photocatalytic Performance. *AIP Conf. Proc.* **2021**, *2339*, 020187.
80. Cha, B.J.; Saqlain, S.; Seo, H.O.; Kim, Y.D. Hydrophilic Surface Modification of TiO_2 to Produce a Highly Sustainable Photocatalyst for Outdoor Air Purification. *Appl. Surf. Sci.* **2019**, *479*, 31–38. [CrossRef]
81. Aravind, M.; Amalanathan, M.; Mary, M.S.M. Synthesis of TiO_2 Nanoparticles by Chemical and Green Synthesis Methods and Their Multifaceted Properties. *SN Appl. Sci.* **2021**, *3*, 1–10. [CrossRef]
82. Nabi, G.; Raza, W.; Tahir, M.B. Green Synthesis of TiO_2 Nanoparticle Using Cinnamon Powder Extract and the Study of Optical Properties. *J. Inorg. Organomet. Polym. Mater.* **2020**, *30*, 1425–1429. [CrossRef]
83. Honarmand, M.M.; Mehr, M.E.; Yarahmadi, M.; Siadati, M.H. Effects of Different Surfactants on Morphology of TiO_2 and Zr-Doped TiO_2 Nanoparticles and Their Applications in MB Dye Photocatalytic Degradation. *SN Appl. Sci.* **2019**, *1*, 505. [CrossRef]
84. Bakardjieva, S.; Šubrt, J.; Štengl, V.; Dianez, M.J.; Sayagues, M.J. Photoactivity of Anatase–Rutile TiO_2 Nanocrystalline Mixtures Obtained by Heat Treatment of Homogeneously Precipitated Anatase. *Appl. Catal. B Environ.* **2005**, *58*, 193–202. [CrossRef]
85. Daneshvar, N.; Salari, D.; Niaei, A.; Rasoulifard, M.H.; Khataee, A.R. Immobilization of TiO_2 Nanopowder on Glass Beads for the Photocatalytic Decolorization of an Azo Dye C.I. Direct Red 23. *J. Environ. Sci. Health Part A* **2005**, *40*, 1605–1617. [CrossRef]
86. Khan, M.I.; Bhatti, K.A.; Qindeel, R.; Althobaiti, H.S.; Alonizan, N. Structural, Electrical and Optical Properties of Multilayer TiO_2 Thin Films Deposited by Sol–Gel Spin Coating. *Results Phys.* **2017**, *7*, 1437–1439. [CrossRef]
87. Guo, M.-Z.; Maury-Ramirez, A.; Poon, C.S. Photocatalytic Activities of Titanium Dioxide Incorporated Architectural Mortars: Effects of Weathering and Activation Light. *Build. Environ.* **2015**, *94*, 395–402. [CrossRef]
88. Loh, K.; Gaylarde, C.C.; Shirakawa, M.A. Photocatalytic Activity of ZnO and TiO_2 'Nanoparticles' for Use in Cement Mixes. *Constr. Build. Mater.* **2018**, *167*, 853–859. [CrossRef]
89. Wu, X. Applications of Titanium Dioxide Materials. In *Titanium Dioxide—Advances and Applications*; IntechOpen: London, UK, 2022.
90. Zulkifly, K.; Heah, C.Y.; Liew, Y.M.; Abdullah, M.M.A.B.; Abdullah, S.F.A. The Synergetic Compressive Strength and Microstructure of Fly Ash and Metakaolin Blend Geopolymer Pastes. *AIP Conf. Proc.* **2018**, *2045*, 020100.
91. Ratan, J.K.; Saini, A. Enhancement of Photocatalytic Activity of Self-Cleaning Cement. *Mater. Lett.* **2019**, *244*, 178–181. [CrossRef]
92. Pathak, S.S.; Vesmawala, G.R. Effect of Nano TiO_2 on Mechanical Properties and Microstructure of Concrete. *Mater. Today Proc.* **2022**, *65*, 1915–1921. [CrossRef]
93. Sun, Y.; Chen, L.; Bao, Y.; Zhang, Y.; Wang, J.; Fu, M.; Wu, J.; Ye, D. The Applications of Morphology Controlled ZnO in Catalysis. *Catalysts* **2016**, *6*, 188. [CrossRef]
94. Fariza, M.; Rashid, A.; Ainuddin, A.R. Zinc Oxide Nanoparticle Synthesize by Green Approach. *Res. Prog. Mech. Manuf. Eng.* **2021**, *2*, 390–398.
95. Garg, N.; White, C.E. Mechanism of Zinc Oxide Retardation in Alkali-Activated Materials: An in-Situ X-Ray Pair Distribution Function Investigation. *J. Mater. Chem. A* **2017**, *5*, 11794–11804. [CrossRef]
96. Nochaiya, T.; Sekine, Y.; Choopun, S.; Chaipanich, A. Microstructure, Characterizations, Functionality and Compressive Strength of Cement-Based Materials Using Zinc Oxide Nanoparticles as an Additive. *J. Alloys Compd.* **2015**, *630*, 1–10. [CrossRef]
97. McKeen, L.W. The Components of Paint. In *Fluorinated Coatings and Finishes Handbook*; Elsevier: Amsterdam, The Netherlands, 2016; pp. 51–58.
98. Goncharenko, D.; Aleinikova, A.; Kabus, O.; Kolomiiets, Y. Study of the Efficiency of Epoxy Coating Protection of Concrete Surfaces from Sulfuric Acid Corrosion. *IOP Conf. Ser. Mater. Sci. Eng.* **2019**, *708*, 012081. [CrossRef]
99. Xiao, Z.; Liu, Y.; Wang, Y.; Shi, J. TA/Fe (III) Anti-Chloride Coating to Protect Concrete. *J. Clean. Prod.* **2020**, *259*, 120922. [CrossRef]

100. Papakonstantinou, C.G.; Balaguru, P.N. Geopolymer protective coatings for concrete. In Proceedings of the SAMPE '07: M and P—From Coast to Coast and Around the World, Conference Proceedings, International SAMPE Symposium and Exhibition, Baltimore, MD, USA, 3–7 June 2007.
101. Pradhan, S.; Pandey, P.; Mohanty, S.; Nayak, S.K. Insight on the Chemistry of Epoxy and Its Curing for Coating Applications: A Detailed Investigation and Future Perspectives. *Polym. Plast. Technol. Eng.* **2016**, *55*, 862–877. [CrossRef]
102. Kozak, A. Application of Acrylic-Based Coatings for Concrete Protection. *MATEC Web Conf.* **2018**, *163*, 05011. [CrossRef]
103. Cong, P.; Cheng, Y. Advances in geopolymer materials: A comprehensive review. *J. Traffic Transp. Eng. (Engl. Ed.)* **2021**, *8*, 283–314. [CrossRef]
104. Jiang, C.; Wang, A.; Bao, X.; Chen, Z.; Ni, T.; Wang, Z. Protective Geopolymer Coatings Containing Multi-Componential Precursors: Preparation and Basic Properties Characterization. *Materials* **2020**, *13*, 3448. [CrossRef] [PubMed]
105. Mao, Y.; Biasetto, L.; Colombo, P. Metakaolin-Based Geopolymer Coatings on Metals by Airbrush Spray Deposition. *J. Coat. Technol. Res.* **2020**, *17*, 991–1002. [CrossRef]
106. Aguirre-Guerrero, A.M.; Robayo-Salazar, R.A.; de Gutiérrez, R.M. A Novel Geopolymer Application: Coatings to Protect Reinforced Concrete against Corrosion. *Appl. Clay Sci.* **2017**, *135*, 437–446. [CrossRef]
107. Sarumathi, M.; Ramaswamy, S.N. Performance and Effectiveness of Concrete Coatings-A State of Art Review. *Int. J. Sci. Res.* **2016**, *5*, 294–298.
108. Rosales, A.; Esquivel, K. SiO_2@TiO_2 Composite Synthesis, and Its Hydrophobic Applications: A Review. *Catalysts* **2020**, *10*, 171. [CrossRef]
109. Guzmán-Aponte, L.A.; de Gutiérrez, R.M.; Maury-Ramírez, A. Metakaolin-Based Geopolymer with Added TiO_2 Particles: Physicomechanical Characteristics. *Coatings* **2017**, *7*, 233. [CrossRef]
110. El Alouani, M.; Alehyen, S.; el Achouri, M.; Taibi, M. Preparation, Characterization, and Application of Metakaolin-Based Geopolymer for Removal of Methylene Blue from Aqueous Solution. *J. Chem.* **2019**, *2019*, 1–14. [CrossRef]
111. Duxson, P.; Provis, J.L.; Lukey, G.C.; van Deventer, J.S.J. The Role of Inorganic Polymer Technology in the Development of 'Green Concrete. ′ *Cem. Concr. Res.* **2007**, *37*, 1590–1597. [CrossRef]
112. Jaya, N.A.; Abdullah, M.M.A.B.; Li, L.-Y.; Sandu, A.V.; Hussin, K.; Ming, L.Y. Durability of Metakaolin Geopolymers with Various Sodium Silicate/Sodium Hydroxide Ratios against Seawater Exposure. *AIP Conf. Proc.* **2017**, *1887*, 020063.
113. Guzmán-Carrillo, H.R.; Manzano-Ramírez, A.; Garcia Lodeiro, I.; Fernández-Jiménez, A. ZnO Nanoparticles for Photocatalytic Application in Alkali-Activated Materials. *Molecules* **2020**, *25*, 5519. [CrossRef] [PubMed]
114. Yaakob, S.M.; Rabat, N.E.; Sufian, S. Effects of Na: Al and Water: Solid Ratios on the Mechanical Properties of Fly Ash Based Geopolymer. *IOP Conf. Ser. Mater. Sci. Eng.* **2018**, *458*, 012011. [CrossRef]
115. Wang, L.; Geddes, D.A.; Walkley, B.; Provis, J.L.; Mechtcherine, V.; Tsang, D.C.W. The Role of Zinc in Metakaolin-Based Geopolymers. *Cem. Concr. Res.* **2020**, *136*, 106194. [CrossRef]
116. Strini, A.; Roviello, G.; Ricciotti, L.; Ferone, C.; Messina, F.; Schiavi, L.; Corsaro, D.; Cioffi, R. TiO_2-Based Photocatalytic Geopolymers for Nitric Oxide Degradation. *Materials* **2016**, *9*, 513. [CrossRef]
117. Saufi, H.; el Alouani, M.; Alehyen, S.; el Achouri, M.; Aride, J.; Taibi, M. Photocatalytic Degradation of Methylene Blue from Aqueous Medium onto Perlite-Based Geopolymer. *Int. J. Chem. Eng.* **2020**, *2020*, 1–7. [CrossRef]
118. Kaya-Özkiper, K.; Uzun, A.; Soyer-Uzun, S. Red Mud- and Metakaolin-Based Geopolymers for Adsorption and Photocatalytic Degradation of Methylene Blue: Towards Self-Cleaning Construction Materials. *J. Clean. Prod.* **2021**, *288*, 125120. [CrossRef]
119. Zhang, Y.; Liu, L. Fly Ash-Based Geopolymer as a Novel Photocatalyst for Degradation of Dye from Wastewater. *Particuology* **2013**, *11*, 353–358. [CrossRef]
120. Chen, L.; Zheng, K.; Liu, Y. Geopolymer-supported photocatalytic TiO_2 film: Preparation and characterization. *Constr. Build. Mater.* **2017**, *151*, 63–70. [CrossRef]