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Review

TiO_2-Based Mortars for Rendering Building Envelopes: A Review of the Surface Finishing for Sustainability

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Abstract: Building envelopes coated with TiO_2-based mortars benefit from depolluting, antibiological and self-cleaning effects. Therefore, photocatalytic renders are allies in the quest for sustainability in the built environment, potentially combatting atmospheric pollution, enhancing durability and reducing maintenance needs. Surface finishing characteristics of the renders influence their photocatalytic efficiency and esthetic and functional properties. In this context, this study reviews the existing literature, focusing on proven surface-affecting parameters, the surface and color of TiO_2-based mortars, to explore their impacts on photoactive behavior. The incorporation of TiO_2 within an additional surface layer and its mixture into the mortar in bulk were observed for surface roughness. Mainly the addition of TiO_2 during casting was identified in colored mortars. Generally, a moderate surface roughness led to better photoactivity; microroughness affected self-cleaning by facilitating dirt deposition. The interaction between the surface roughness and the photocatalytic layer affected the water contact angle, regarding superhydrophilicity or superhydrophobicity. The photoactivity of colored mortars with TiO_2 depended on the color and amount of the added pigments, which influenced electron–hole recombination, physically occupied active sites or, on the other hand, led to a higher formation of reactive radicals. Surface finishing can thus be designed to enhance the photoactivity of TiO_2-based mortars, which is fundamental for current climate concerns and emphasizes the need for life cycle assessments and environmental protection.

Keywords: TiO_2; mortar; surface finishing; roughness; pigment; color; building envelope; sustainability

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1. Introduction

TiO_2 is the most researched photocatalytic material in the construction field, and its use in mortars has been recommended as a strategy to improve the performance of building renderings seeking to obtain self-cleaning surfaces [1,2]. The massive implementation of mortars with TiO_2 in construction could also significantly reduce air pollution as the photocatalyst is known to oxidize airborne organic and inorganic compounds, degrade biological pollutants and inhibit microorganisms' growth [1,3]. Therefore, TiO_2 in mortars fosters sustainability and environmental protection by combatting atmospheric pollution in more durable façades with lower maintenance needs [4]. The compatibility of TiO_2 with construction materials [5] and the large outdoor area provided by the built environment [6] reinforce the photocatalyst's potential. In addition, buildings account for 36% of the energy-related greenhouse gas emissions and 40% of the total energy consumed [7], urging for façades that actually fight climate challenges and, thus, justifying the research of TiO_2-based renderings. Therefore, many researchers, companies and practitioners have investigated the effect of TiO_2-based mortars aiming to optimize the development of the coatings [1].

TiO_2 exists in three principal varieties of crystalline structures: anatase, rutile and brookite [8], which lead to differences in photocatalytic efficiency, with anatase being the most photoactive polymorph [9,10]. Anatase absorbs ultraviolet radiation (UV) with a wavelength lower than 380 nm [11] and then works through heterogeneous photocatalysis, an advanced oxidative process (AOP) associated with substantially reactive hydroxyl (OH•) radicals [12].

Heterogeneous photocatalysis involves the activation of a semiconductor, such as TiO_2, by natural or artificial light; the bandgap energy to be surpassed between TiO_2 valence (VB) and conduction (CB) bands seeking activation is 3.2 eV [13]. Thus, when photons with higher energy than the TiO_2 bandgap are absorbed, electrons (e^-) may transfer from the VB to the CB, forming highly oxidant holes (h^+) in the VB, which may further react with water molecules releasing OH• [14,15]. OH• radicals and h^+ may coexist, promoting oxidation reactions on the photocatalyst surface [16].

Although the oxidative processes are prevalent due to the high potential of hydroxyl radicals and holes, reduction reactions may also contribute to the photocatalytic process [17]. Superoxide anions and hydroperoxide radicals result from the e^- transfer during photocatalysis [18] and can act in mineralizing pollutants as well [19].

Superhydrophilicity is also photoinduced by TiO_2, mainly regarding the availability of holes [20]. It is the synergy between the photocatalytic reactions involving the oxidation and reduction of adsorbed substances with the superhydrophilicity that substantiates TiO_2 use in construction materials seeking water and air purification, self-cleaning behavior and anti-bacterial effects [21]. Thus, TiO_2 is an ally in tackling key ecological challenges, like environmental cleanup, using solar energy [22] and addressing major concerns about urban buildings' energy consumption and air quality [5].

Wei et al. [5] studied TiO_2-based materials aiming to support green, low-carbon buildings and sustainable urban growth. In fact, the benefits of incorporating TiO_2 in buildings can even be assessed from the climate change perspective since its mitigation mainly depends on reducing the emission of greenhouse gases [23]. TiO_2 has been studied not only for its ability to reduce CO_2 [24,25] but also its effect on air purification concerning other pollutants, such as nitrogen oxides (NO_x) [26], and the degradation of several organic contaminants [22]. Indeed, the parameters of the façades affect embodied and operational impacts during the life cycle of buildings [27]. The effect of TiO_2 towards sustainability in construction should be further investigated and quantified, for instance, by encompassing the reductions in the carbon footprint due to the degraded gases, usually measured through their carbon dioxide equivalent [28]. Unfortunately, knowledge about the life cycle assessment of TiO_2 coatings still lacks information [29], and it should be addressed, particularly the existing concerns on the ecotoxicity of nanomaterials and their impact on human health and the environment [30].

The incorporation of TiO_2 in mortars can be done in the pre-casting phase for dry photocatalytic mortar mixtures [31], during casting by adding the nanoparticles to the other materials while mixing [32], or in the post-casting phase as an additional surface layer [33]. In Krishnan et al. [34], similar photoactivity was obtained by comparing silicate coatings containing TiO_2 and the incorporation of a semiconductor within the mortar mixture; however, the silicate coating required 20 times less TiO_2 [34] since only the surface layer is actually activated by UV radiation [33]. On the other hand, bulk admixtures show higher stability regarding surface erosion, even though they may waste potentially active material in their depth [33]. The optimum ratio of TiO_2 is debated in the literature, and it changes significantly regarding the photocatalyst incorporation method [34]. Furthermore, concerning TiO_2-based mortars and their compressive strength, for instance, adding more photocatalyst than the optimum content may decrease the arising results [35]. Therefore, a trade-off exists when considering the benefits and concerns of the different TiO_2 integrating methods in mortars.

The advantages of incorporating TiO_2 nanoparticles could be extended to the durability increase of the surface and reduction of building cooling demands [36,37]. Reductions in the energy consumption of buildings are essential to decrease their environmental footprint, and green-certified buildings have been proven to mitigate greenhouse gas emissions, further contributing to human health [38]. This could be achieved by improving UV protection [39] and increasing the near-infrared (NIR) reflectance of colored building envelope materials [40] and dark coatings [41].

When the esthetic value is expected from the surface of mortars, color variations [42] and damages, like stains and leaching, become especially relevant [43]. This is the case with TiO_2-based mortars, which do not receive additional outer layers since they could represent a barrier for UV radiation penetration and, thus, prevent photoactivation [44]. Moreover, roughness has been reported as a factor influencing a series of characteristics in mortars, such as wettability [45]. Also, the coating thickness was identified, through finite-difference time-domain (FDTD) numerical simulations, as a dependent factor of the coating reflectance [46]. Color, reflectance, emissivity, liquid water permeability [47,48], gloss, roughness, compactness and hardness are among the surface properties investigated as solutions to be applied to building façades [49–51].

In this context, Dantas et al. [52], for example, evaluated how different scales of surface roughness affect the activation of TiO_2 in mortars. Hot et al. [53] also mentioned the potential of controlling substrate roughness, seeking optimal photocatalytic efficiency. Hamidi and Aslani [54] referred to reports on the influence of surface roughness on the performance of photocatalytic cementitious materials. However, most of the research on photocatalytic TiO_2-based mortars relates to white finishing substrates [55], even though colorful products are increasingly demanded [56] and may be attractive for design specifications. The interest in photocatalytic colored renders may be emphasized regarding the fact that biocolonization [57] and soot deposition [10] lead to changes in the surface color, which may be addressed by TiO_2 photoactivation.

Therefore, to fully explore the potential of TiO_2-based mortars in the current quest for sustainability and durability in the built environment, the present paper discusses surface finishing characteristics that may be controlled and can impact photocatalytic efficiency, such as the resulting self-cleaning activity and maintenance needs, its ability to fight against air pollution and the potential action against microorganisms. Due to their already reported influence on TiO_2 photoactivity in mortars, the focus is dedicated to surface roughness and color, and a detailed overview has been developed, based on existing research, to highlight trends and opportunities for the development of photocatalytic renderings during the current climate change and global warming concerns. The importance of studies seeking the best approach involving the choice of materials and design for building façades is evident.

The methodology used in this paper to achieve a detailed review was primarily based on the Scopus and Web of Science databases. After assessing the main documents retrieved from searches using pertinent query strings and commenting on their results, further discussions were made based on the related existing literature. The first section of the paper presents research studies that discuss the implications of changing surface roughness in TiO_2-based mortars, and the second one presents a detailed overview of the studies that focus on adding colored pigments to photocatalytic cement-based materials with TiO_2.

2. Surface Roughness in TiO_2-Based Mortars

2.1. Overview of the Existing Research

The surface roughness influence of photocatalytic mortars was examined by searching in the Scopus database within the title, abstract and keywords of scientific papers in English, using the terms "mortar AND (titanium AND dioxide OR TiO_2) AND photocatal* AND rough*". This resulted in 15 documents, of which 11 were journal papers and four were conference abstracts. An additional search was performed using the same terms in the Web of Science database within all fields, resulting in two other journal papers (after eliminating the double results). Table 1 presents a summary of the identified documents for this section.

Table 1. Summary of the identified surface roughness-related documents.

Paper	Surface Roughness Treatment	Substrate Compositions	Type of TiO_2 Addition
Giannantonio et al. [58]	Surfaces brushed and polished (with 120 or 600 grit)	Different combinations of cement (Holcim GU I/II cement, Holcim GU + limestone, Essroc I/II, or Essroc I/II + TiO_2); water-to-cement ratio (0.3, 0.4, or 0.6); supplementary cementitious materials (fly ash, slag, silica fume or metakaolin, all in different percentages)	Small percentage of nano-anatase TiO_2 added to the cement
Zhang et al. [59]	Surface finish created using a 3:1 sand-to-cement ratio for all tested mortars, which received different types of emulsions (zinc oxide- or titanium dioxide-based), after 10 days of cure	3:1 sand-to-cement with 0.6 water/cement ratio	<0.1 (wt%) of nano-TiO_2 incorporation in emulsion
Radulovic et al. [60]	3:1 (sand:cement) ratio for all mortars, which received different types of TiO_2-based emulsions, after 31 days of cure	3:1 sand-to-cement with 0.6 water/cement ratio (similar to Zhang et al. [59])	Incorporation of anatase nano-TiO_2 in emulsions at different percentages (<0.1, <0.3, or <0.5 wt%)
Hot et al. [53]	Obtained by using two formulations of mortars: with a water–cement ratio of 0.4 for higher roughness and of 0.5 for lower roughness	Based on Portland cement and siliceous sand, with a formulation based on NF EN 196-1 standard [61]	TiO_2 dispersion applied as a coating to the substrate surface using a fine brush in two layers (dry matter contents of TiO_2 in wt% were 18, 12, 6, 5, 4, 3 and 1)
Yang et al. [62]	Different surface roughness developed by producing mortars with ordinary Portland cement (OPC) or alkali-activated slag (AAS), receiving or not TiO_2-based coatings	OPC mortars were prepared in a mass proportion of 1:0.5:3 (cement, water and sand); AAS mortars had a mass ratio of 1:0.4:2 (slag, water and sand)	A TiO2@CoAl-LDH suspension (created using nanospheres of TiO_2 and CoAl-layered double hydroxide shell) was sprayed as a double coating layer in the mortar surfaces
Ruot et al. [63]	Siliceous sand increased the mortar's roughness when compared to cement paste	Mortars were developed using 65 wt% siliceous sand, 30 wt% CEM I Portland cement, 5 wt% calcareous filler (99.3% $CaCO_3$) and additions of 0.5 wt% of hydroxyethylmethyl cellulose and different percentages of nano-TiO_2 in an overall water/cement ratio (wt./wt.) of 1	0, 1, 3 and 5 wt% of anatase nano-TiO_2 (concerning the dry mixture) were added to the mortar mixes
Maiti et al. [64]	Nano-TiO_2 addition to mortars produced less roughness in the surface samples when compared to the reference mortar	Geopolymer mortars prepared by mixing fly ash (FA), alkali activator (AA; 10M NaOH and Na_2SiO_3 as 1:2 ratios), normal sands (S) and different sizes of nano-TiO_2	5% (*w*/*w*) of TiO_2 nanoparticles were added to the mortar mix in different particle sizes (30 nm, 50 nm and 100 nm)

Table 1. *Cont.*

Paper	Surface Roughness Treatment	Substrate Compositions	Type of TiO_2 Addition
Naganna et al. [65]	Roughness decreased by the addition of TiO_2	Mortars were prepared using ordinary Portland cement, river sand and nano-TiO_2 in a binder to fine aggregate ratio of 1:3 (by weight). Water was added at percentages between 9.5 and 11.25% (of the total mass of binder and sand)	Cement was replaced with a rutile nano-TiO_2 at quantities of 0%, 2%, 4%, 6%, 8% and 10% by weight
Dantas et al. [52]	Roughness was set to vary according to the type of TiO_2 used	Different compositions using white Portland cement, dolomite, polypropylene microfibers and TiO_2	Commercial TiO_2 (P25 and P105, mostly in anatase phase) in powder and in suspensions were added to the cement matrix
Zahabizadeh et al. [66]	Surface roughness of 3D printed cementitious materials is supposed to be higher because of the lack of mold's walls	Cement CEM I 42.5 R, fly ash type F, silica fume, fine quartz sand, a polycarboxylate-based superplasticizer and water	Nano-TiO_2 (80% anatase) aqueous suspensions with a pH of 8 (in rates of 4, 8 and 16 g/L) were applied as a coating to the 3D-printed mortars
Vulic et al. [67]	The nano coating improved the hydrophilicity and decreased the surface roughness	Different compositions, with a group using Portland cement, water and sand in the ratio of 1:0.5:3; and another group using pozzolanic material and lime ($Ca(OH)_2$) in a mass ratio 2:1, with a sufficient amount of water for a specific consistency	TiO_2/ZnAl layered double hydroxides were applied as a coating to the prepared mortars
Casagrande et al. [68]	Roughness varied according to the type of mortar used	A ready-to-mix industrialized mortar and a mortar produced in laboratory were used, both containing Portland cement, sand, admixture and water	Three types of commercial nano-TiO_2 in powder (mostly composed by anatase) were mixed in proportions of 3, 5, 7 and 10 wt% (relative to cement weight)
Graziani et al. [69]	Specimens produced with different total porosity and roughness	Substrates were made of two different types of clay brick: the first one using a high porosity method (to replicate Cultural Heritage bricks), and the second using a modern technique to produce low porous brick	Two types of aqueous TiO_2-based solutions were applied as spray coatings (one doped with silver nanoparticles and the other one with copper nanoparticles)

Concerning the surface roughness, the study from Casagrande et al. [68], which analyzed the fresh and hardened properties of nano-TiO_2-added cement mortars, presented different results depending on the type of mortar: the nanoparticles led to a smoother surface in industrialized mortars, whereas in nonindustrialized mortars, they possibly led to a rougher surface.

Within all the identified documents in the databases, three focused on the biologic growth analysis of photocatalytic mortars, such as the work of Giannantonio et al. [58], who used different fungal strains to evaluate the effects of several concrete characteristics (including different cement compositions, material additions and surface roughness) in biofouling and fungal colonization. The changes in surface roughness of the mortar tiles could not statistically relate to different biofouling propensities, although only three distinct surface roughness types were tested (a standard brush finish, a 120-grit paper polished finish and a 600-grit paper polished finish, and two of them had a very similar smoothness) [58].

The results of Giannantonio et al. [58] can be related to the findings of another paper—the study from Zhang et al. [59] identified that the change in surface roughness was not a factor altering bioreceptivity. In this case, emulsions containing either TiO_2 or zinc oxide (ZnO) were applied as surface treatments in a 3:1 sand–cement mortar (by using a pre-wetted brush) and resulted in similar average values, which was evaluated using a Mitutoyo CV500 contour measuring instrument [59] (Mitutoyo America Corporation, Aurora, IL, USA). The same equipment was used by Radulovic et al. [60], whose work analyzed the biofouling resistance of façade treatments with incorporated nano-TiO_2. Again, the surface roughness did not influence the algal colonization since all tested specimens had relatively similar values for porosity and surface roughness [60]. The work from Graziani et al. [69] was the only one from the retrieved papers that did not use mortar as a substrate or matrix for interaction with TiO_2. The authors applied a silver and copper nano-particulate enhanced with an aqueous nano-titania solution on bricks with different porosities and roughness and found that algal growth was minimized in low-porous bricks covered with the solution. Although greater surface roughness has been reported as having a strong influence on increasing biological colonization [70,71], some studies, where different surface treatments with the addition of TiO_2, were performed, could not establish a clear relationship between the two factors [58–60].

Concerning the depolluting properties of TiO_2-based mortars, Hot et al. [53] analyzed the degradation of nitric oxide (NO) by photocatalytic oxidation in mortars of different surface roughness created by two water/cement ratios (0.4 for higher roughness and 0.5 for lower values) coated with TiO_2 photocatalytic dispersions by brushing. The optimal photocatalytic performance was achieved using a substrate of moderate roughness with a controlled amount of TiO_2 on the surface. Although Hot et al. [53] suggested further investigation on surface roughness characterization, in their study with photocatalytic dispersion coatings, the type of substrate had a significant impact on the adhesion of TiO_2 particles—with rougher surfaces accommodating a more significant amount of TiO_2 particles—and a potential reduction was identified in the NO degradation rate on excessively rough surfaces since particles were less exposed to light [53].

In this context, Yang et al. [62] assessed the depolluting effect using methylene blue photocatalytic removal in ordinary Portland cement and alkali-activated slag-added mortars coated with a TiO_2@CoAl-LDH suspension and observed the relationship with a variety of properties, including roughness and microhardness. The photocatalytic coating included innovative core–shell nanospheres with a TiO_2 core and a CoAl-layered double hydroxide shell. It was responsible for a roughness decrease through its filling effect, which lowered the likelihood of biofouling on the surface, thus enabling a self-cleaning capability [62]. Furthermore, Yang et al. [62] evaluated the effects of rainwater washing on the surfaces and found that mortars with alkali-activated compositions were more susceptible to presenting a reduced surface roughness after rain, resulting in a higher flux of organic pollutants and increased availability of photocatalytic active sites, consequently resulting in enhanced photoactivity. Also, regarding the removal of a dye (Rhodamine–B—RhB) as an indicator for photocatalytic efficiency, Ruot et al. [63] found that TiO_2-based mortar specimens presented a higher surface roughness than cement pastes (due to the presence of sand), and the formulation with 3 wt% of anatase showed higher surface content of TiO_2 concerning the addition of 1 to 5 wt%, which could be due to its high porosity.

The works that evaluated surface roughness in construction materials focused especially on cementitious substrates [53,58–60,62]. However, Maiti et al. [64] performed rutile nano-TiO_2 incorporation in different geopolymeric composites. One of the main analyses using mercury intrusion porosimetry (MIP) and atomic force microscopy (AFM) revealed that 30 nm of TiO_2 nanoparticles decreased both the porosity and surface roughness in the structure, which resulted in a denser matrix and enhanced the overall durability performance [64]. In the same context, durability and strength were evaluated in the study by Naganna et al. [65] regarding the potential of incorporating nano-TiO_2 particles into cement to create mortar and concrete with reduced permeability. In their findings, adding nano-TiO_2 into concrete specimens significantly mitigated the roughness and surface defects of coarse aggregates and improved the strength and durability characteristics (when added as a substitution of cement in quantities up to 8%) [65].

2.2. Methods and Tests to Evaluate Surface Roughness

The methods to evaluate surface roughness retrieved from the literature include quantification of surface roughness by using a Leica SP-1 Confocal Microscope [58] (Leica Microsystems, Wetzlar, Germany), which can produce a roughness number. This was previously described in the works of Kurtis et al. [72] and Chinga et al. [73]. Atomic force microscopy (AFM) with a silicon tip cantilever, employing the Dimension Icon system from Bruker [62], was also used. Maiti et al. [64] also applied AFM to evaluate the roughness and morphology of the geopolymer specimens. Hot et al. [53] applied a method involving an initial surface measurement campaign with a confocal laser scanning microscope (Olympus LEXT OLS3100, Olympus Global, Tokyo, Japan) on each studied substrate. In the referred method [53], scans were conducted across the specimen's field of view, using a photomultiplier tube to convert light into electricity. By collecting a series of 2D digital images at various depths, 3D images were generated, allowing for the determination of roughness values.

Following a different approach to measure surface roughness, Dantas et al. [52] used an optical profilometer (as a noncontact method), scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) elemental mapping to characterize mortar surfaces that received different treatments of TiO_2—either in powder (added in the cement matrix) or in suspension (commercial solutions and suspensions prepared by the authors). The results showed that microroughness does not directly affect TiO_2 activation but can enhance dirt adhesion, potentially hindering TiO_2 activation. The presence of various roughness levels and the use of the same TiO_2 particle size can influence the dispersion of the photocatalytic product on surfaces [52]. Moreover, TiO_2 activation depends not solely on direct solar radiation but also on diffuse radiation, which mitigates microshading due to peaks and valleys [52].

Zahabizadeh et al. [66] also applied SEM and EDS to evaluate the surface roughness of 3D-printed photocatalytic mortars, with the addition of X-ray diffraction (XRD) and a MICROTOP.06.MFC device, which served as a nondestructive, noncontact method designed for analyzing the microtopography of small-sized samples in 2D/3D [74]. The surface analysis of the 3D-printed specimens revealed a negative surface texture for those with significant gaps between irregularities, making them conducive for semiconductor particle deposition. Additionally, the specimen treated with an 80 mg/cm^2 nano-TiO_2 aqueous solution exhibited the highest photocatalytic efficiency, indicating the evenest distribution of TiO_2 nanoparticles on the coated surface [66]. Table 2 summarizes the test methods used in all the collected studies.

Table 2. Summary of the identified test methods to evaluate surface roughness.

Paper	Method/Test for Roughness Measurement	Parameters Obtained from the Method
Giannatonio et al. [58]; Kurtis et al. [72]	Quantification of surface roughness using laser scanning confocal microscopy	Roughness number obtained with a methodology that uses images from confocal microscopy
Yang et al. [62]; Maiti et al. [64]	Atomic force microscopy (AFM)	R_a parameter, which indicates the average roughness value. A resulting 3D image also provides morphology parameters
Hot et al. [53]	Confocal laser scanning microscope	Determination of roughness value from 3D images generated with a series of 2D digital images at various depths
Dantas et al. [52]	Optical surface topography (profilometer)	Shape and structure analyzed by the creation of Gaussian distributions, linearly combined, expressing results in terms of probability density
Zahabizadeh et al. [66]	X-ray diffraction (XRD)	Crystal structure (used for comparison of different crystallographic structures)
Dantas et al. [52]; Zahabizadeh et al. [66]; Casagrande et al. [68]	Scanning electron microscopy (SEM) (with energy-dispersive X-ray Spectroscopy (EDS) elemental mapping)	SEM analyzes surface morphology providing particle dimension parameters, combined with EDS to evaluate TiO_2 dispersion
Zahabizadeh et al. [66]; Costa et al. [74]	Analysis of microtopography using a MICROTOP.06.MFC device	Average roughness (S_a), Root-mean-square deviation (S_q), total roughness height (S_t), maximum peak height (S_p), maximum valley depth (S_v), space between the peaks or irregularities (S_m), skewness (S_{sk}), kurtosis (Sk_u), area of the peak portion (S_{r1}) and area of the valley portion (S_{r2})
Graziani et al. [69]	Portable rugosimeter	Arithmetic average (R_a), maximum profile peak height (R_z) and maximum height of the profile (R_{max}), calculated according to the UNI EN ISO 4287:2009 [75]
Casagrande et al. [68]	Surface macrotexture analysis adapted from ASTM E-965 [76]	Sand height (SH)

2.3. Evaluation of Hydrophilicity/Hydrophobicity in Photocatalytic Mortars

Most of the collected research showed that lower surface roughness values are usually related to higher hydrophilicity in TiO_2-based or coated specimens [62,67]. For example, in the study of Yang et al. [62], the photoinduced hydrophilicity related to the reduction in the contact angle is suggested as the primary factor behind the rise in surface water absorption in mortars coated with TiO_2 and a CoAl-layered double hydroxide shell under ultraviolet–visible irradiation. Vulic et al. [67] analyzed the photoinduced characteristics after repeated wetting of the mortar surface and ultraviolet–visible light exposure and found that the decline in the initial contact angle and the rise in surface water absorption in the coated mortars exposed to UV light also resulted from the photoinduced hydrophilicity effect.

On the other hand, in the work of Radulovic et al. [60], TiO_2 treatments enhanced silane/siloxane water-repellent emulsions, keeping superhydrophobic properties that possibly persisted under varying external conditions, potentially ensuring that internal pores remained superhydrophobic. This could offer dual benefits of protecting the substrate from rising damp and biofouling. Similarly, in the work of Zhang et al. [59], the treatments with ZnO and TiO_2 achieved slightly higher water contact angles compared to the control emulsion, which is connected to a hydrophobic trend.

2.4. Final Remarks

Overall, the research on TiO_2-based mortars indicated that the addition of the photocatalyst decreases surface roughness due to its filler effect and that mainly moderate mortar surfaces may improve the efficiency, strength and durability performance. A summary of the principal results of the evaluated works is presented in Figure 1. Microroughness was reported as not directly affecting TiO_2 activation but as enhancing dirt adhesion, potentially holding back TiO_2 activation. Moreover, both hydrophilic and hydrophobic properties were observed in different approaches for changing surface roughness in TiO_2-based mortars. Indeed, it is the interaction between the photocatalytic layer and the surface that defines its performance regarding the hydrophobicity, surface roughness and durability [77]. Since, among others, different NO_x removal efficiencies [78] and, therefore, varied responses to atmospheric pollutants derive from surface roughness, this can be considered a relevant parameter influencing the very contribution of TiO_2-based mortars towards sustainability. Further studies should be developed in this area, especially regarding biological growth.

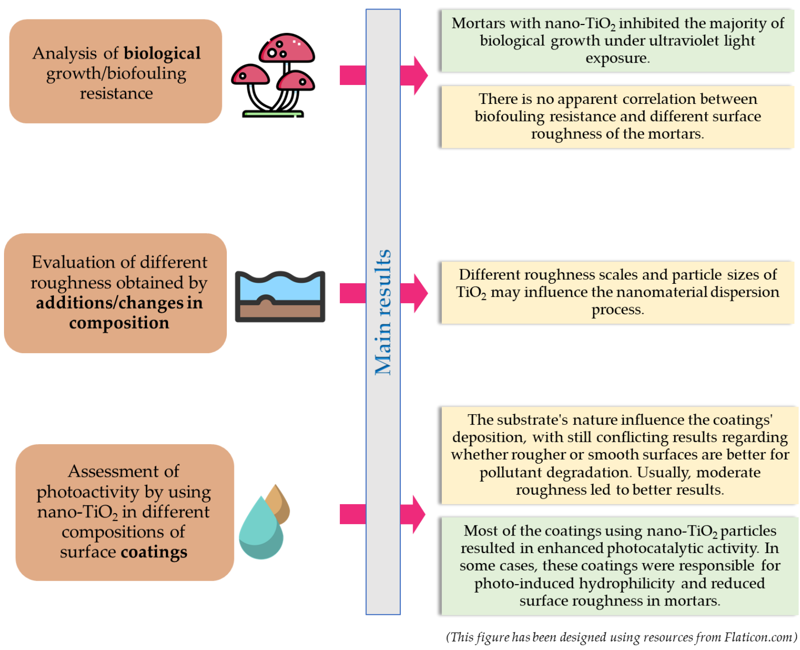

Figure 1. Main topics and results regarding the surface roughness of TiO_2-based mortars based on the literature review.

3. TiO_2-Based Pigmented Mortars

3.1. Overview of Pigments in Mortars

Pigments can be classified as organic or inorganic, aside from colored, white and black [79]. Inorganic pigments are mostly metal oxides, such as carbon black, iron oxides and TiO_2 itself [80]. Indeed, ASTM C979/C979M-16 [81] presents as typical pigments for coloring concrete, some natural or synthetic iron oxides (yellow, red, brown and black), chromium oxide, cobalt blue, carbon black and titanium dioxide. Dantas et al. [52] referred to TiO_2 as pigments in the context of their ability to absorb UV radiation and scatter visible light; in fact, cool pigments are mainly characterized by their solar spectral backscattering and absorption coefficients [82]. Cool pigments may fit within the inorganic category, with the special function of minimizing solar heat accumulation on building surfaces and, thus, decreasing cooling demands [83]. Inorganic pigments should be preferred for coexisting with TiO_2 photocatalysis [33].

Organic pigments consist of organic compounds not soluble in water, such as indigo and Tyrian purple or copper phthalocyanines [84]. Additionally, thermochromic pigments, which reversibly change their color depending on the temperature [85], present three components (typically, cyclic esters, weak acids and alcohol or other nonvolatile hydrophobic compounds) involved with organic microcapsules [86]. Table 3 summarizes some of the main impacts resulting from the addition of several pigments to non-photocatalytic mortars as a basis for the following discussion regarding their joint addition with TiO_2.

Table 3. Pigments and their impacts in non-photocatalytic mortars.

Pigment	Color	Chemical Composition of the Pigment	Paper	Pigment Content	Mortar Composition	Impact of the Pigment on the Mortar
Iron oxide	Yellow	FeOOH [33]	Jang et al. [87]	9% and 12% of cement mass	White Portland cement, granulated ground blast furnace slag, pigment, sand, water	Higher reduction in fluidity than red and green pigments. Yellow pigment has the highest specific surface area and needle-shaped particles
			Hatami and Jamshidi [88]	5% of cement content as a replacement of filler	Cement, water, sand, filler, superplasticizer, pigment	The pigment may act as a filler, preventing reductions in the mechanical characteristics of self-compacting mortars
	Red	Fe_2O_3 [33]	Assaad et al. [89]	2 to 8% of cement mass, at 2% increment rates	Cement, pigment, styrene–butadiene rubber latex, sand, water	Lower rheological properties (yield stress and plastic viscosity) than with yellow pigment. Red pigments are spherical
	Black	Fe_3O_4 [33]	Lee et al. [90]	3 to 12% of cement mass, at 3% increment rates	Cement, sand, pigment, water	Black pigments did not modify flowability up to 9% of the mixing ratio. Black pigments are spherical
	Brown	Mixture of FeOOH, Fe_2O_3, Fe_3O_4 [33]	Lee et al. [91]	3%, 4% and 5% of cement mass	Cement, large size sand, stone dust, medium size sand, pigment, water (concrete interlocking blocks)	4% of brown iron oxide pigment led to the highest flexural strength (concrete interlocking blocks)

Table 3. *Cont.*

Pigment	Color	Chemical Composition of the Pigment	Paper	Pigment Content	Mortar Composition	Impact of the Pigment on the Mortar
Chrome oxide	Green	Cr_2O_3 [87]	Heerah et al. [92]	1%, 5% and 10% of cement mass	Cement, sand, water, pigment	Increasing contents of pigment led to sharp increases in water absorption, porosity, and compressive strength due to impurities (FeO(OH) and mica (muscovite))
Cobaltous aluminate	Blue	$CoO.Al_2O_3$ [92]	Heerah et al. [92]	1%, 5% and 10% of cement mass	Cement, sand, water, pigment	No significant changes in water absorption and porosity, but increasing compressive strength
Thermochromic	Black	pH-sensitive color former, electron-accepting color developer and hydrophobic nonvolatile solvent, enclosed in melamine formaldehyde microcapsules [85]	Perez et al. [85]	3% weight of solid of pigment (slurry = 50% of capsules in aqueous solution)	Cement, calcareous sand, limestone filler, siliceous sand, water repellent, water retainer, resin, fiber, pozzolan, thermochromic slurry, water	High water retention, low apparent density and elasticity modulus. Reduction of 20.5% and 16% in compressive and flexure strength, respectively
	Blue	Crystal violet lactone: 4,4′-dihydroxydiphenyl propane:solvents (phenol, alcohol) = 1:5:30 + terephtaloyl chloride, emulsifying agents, $NaHCO_3$ [93]	Ma and Zhu [93]	Minimum amount of 10% by cement weight	Cement, water, pigment	13% higher water content in the mixture and 20% to 40% lower mechanical strength. Setting time and matrix integrity are not affected

Iron oxide pigments are the most used in construction [94,95]. Indeed, their incorporation in cementitious materials seeking architectonic and decorative purposes is recurrent [89], even though challenges remain regarding their application, especially concerning their color stability throughout time [43]. The ability of the pigments to color cementitious matrices and to impact their mechanical properties rely on their dimension and specific surface [88]. As shown in Table 3, besides impacting flowability, iron oxide pigments affect the strength, shrinkage and durability of the cement composites, mainly influenced by the particulate's format for each pigment color [90].

Studying yellow and red iron oxide pigments, Assaad et al. [89] observed higher rheological properties with increasing contents due to a higher packing density and interparticle links in the matrix. Heerah et al. [92] identified an important problem concerning cobaltous aluminate green pigments, which had impurities within their composition that affected the mortars' performance; therefore, the necessity of the previous characterization of the pigments must be highlighted.

Regarding the pigment contents added to mortars, according to López et al. [43], a saturation and lightness limit exists, above which the surface aspect cannot be more vivid, lighter or darker. The regulation of ASTM C979-C979M-16 [81], which addresses colored concrete, recommends pigment addition contents of ½% and 6%, concerning the cement mass, for preparing mortar specimens.

López-Rebollo et al. [96] pursued energy-efficient mortars by adding white and black commercial pigments composed of synthetic metal oxides, identifying significant increases in temperature for black mortars and only small reductions with the white pigments. Specifically, infrared (IR) reflecting colorants are being researched for surface coating applications, as they reflect the heat radiation to the atmosphere instead of conducting it into the buildings [97]. In the work of Rosso et al. [98], TiO_2 pigments were added as

IR pigments since they are almost white, enhancing thermal–optical characteristics and "coolness". Cool pigments may mitigate urban heat islands [98], increase reflection and reduce energy consumption [99] and could be further obtained by combining iron oxide with TiO_2 [100].

Thermochromic pigments and renderings are especially promising for regions with severe winters and hot summers, leading to a relevant energy economy [101,102]. The thermochromic reaction process is related to a transition temperature, above which the colored state prevails and below which the pigment appears with a colorless aspect [86]. Therefore, regarding colored mortars, pigments may be added for esthetic characteristics, but more than that, they can be instruments to address current climate worries, including global warming and energy efficiency.

3.2. The Interaction of Pigments and TiO_2 in Mortars

Concerning TiO_2-based photocatalytic colored mortars, a search on the Scopus database for the query string "mortar AND (titanium AND dioxide OR TiO_2) AND photocatal* AND pigment" within the title, abstract and keywords of scientific papers in English retrieved four results [31,33,55,103]. In the Web of Science, the query string returned eight journal articles; four of them were already gathered in Scopus, and the other four were from Dantas et al. [52], Guo and Poon [104], Stefanakis et al. [105] and Sugrañez et al. [106]. To expand the search and check its coverage, an additional round comprising concretes with the query string "concrete AND (titanium AND dioxide OR TiO_2) AND photocatal* AND pigment" was carried out, resulting in two papers in English from journals in Scopus (Guo and Poon [104] and Pal et al. [107]) and three papers in the Web of Science, two of which were already identified for mortars [105,106]. From this search, the study of Bahreini et al. [108] was added. Therefore, the assessment of the interaction of pigments and TiO_2 in mortars using Scopus and Web of Science databases comprised initially ten journal papers. However, Bahreini et al. [108] worked mainly with glass plates as reactor walls, not exactly referring to mortars or building envelopes.

Diamanti et al. [33] produced white cement mortars containing glass fiber and 5%anatase; gray, yellow, red and brown pigments were added to the mortars in 2% and 4% contents. Regarding pollutants mineralization, such as volatile organic compounds (VOCs), the photocatalytic pigmented mortars had an efficiency 40% lower than the nonpigmented ones, regardless of the color, due to the interaction among the pigments and the photocatalyst, which was responsible for the higher recombination of electron–hole pairs [33]. Nonpigmented mortars produced by Bersch et al. [55] with 5% and 10% TiO_2 by cement mass showed the best RhB degradation results, irrespective of the photocatalyst content compared with mortars with 4% yellow and brown iron oxide pigments, for which the efficiency increased with higher photocatalyst additions.

Guo and Poon [104], studying the addition of iron oxide pigments to concretes with TiO_2, also observed a reduction in the NO-removal rate in the presence of orange, red, yellow and black pigments, as well as of Cr_2O_3-based green pigment, ranging from 8% to 46%, with the highest impact deriving from the black pigments. In this case, the explanation for the negative effect of the pigments on the photoactivity was due to the occupation of the TiO_2 active sites by the pigments and blocking of surface pores, impacting the NO diffusion; darker pigments could further harm NO removal because of their low scattering potential and higher absorption through the solar light spectrum [104].

On the other hand, for Laplaza et al. [31], who studied a commercial photocatalytic mortar, red and light gray pigmented specimens showed higher NO_x degradation under ultraviolet–visible irradiation than the nonpigmented formulation and the yellow, brown and dark gray mortars. In fact, for fine surfaces, red and light gray photocatalytic mortars promoted nearly 25% and 20% of NO_x degradation, compared to around 13% of TiO_2-containing nonpigmented specimens. Red and light gray photocatalytic mortars led to a higher formation of $OH^{\bullet}$: the electron transfer between the pigments' CB and TiO_2 and vice versa and the Fe/Ti ratio were then identified as influencing parameters [31]. The

Fe/Ti ratio impacted electron–hole recombination since only ratios lower than 0.35 could enhance photoactivity compared to TiO_2 alone [31].

Regarding the production of specimens, Diamanti et al. [33], Guo and Poon [104] and Laplaza et al. [31] separately mixed the iron oxide pigments with the other materials of the photocatalytic cement composites without initially blending them with the TiO_2. On the other hand, Aranzabe et al. [103] first modified the ultramarine blue inorganic pigment with a TiO_2 sol–gel coating around its surface and further studied influencing parameters on the added coating formation. The pH, Ti weight content and the addition of anatase nanoparticles were optimized. Then, after adding 7% in mass of the modified pigments to the mortars, their mechanical properties were investigated, indicating increases in flexural and similar compressive strengths due to the interaction of the pigment with a high specific area with the mortar [103]. Sugrañez et al. [106] proposed new photocatalytic materials derived from industrial sand-blasting operation wastes, which after thermal transformation into α-Fe_2O_3, enabled the photochemical degradation of organic methylene blue dye. The joint use of α-Fe_2O_3 with TiO_2 in cement-based mortars further enhanced photocatalytic activity, contributing to the reduction of the needed amount of TiO_2 in building materials [106].

3.3. Future Trends and Opportunities

Wang et al. [109] reported that TiO_2 doping may be possible with metals, such as Fe^{3+}, seeking to enhance the light absorption spectra to include visible light, for instance, and also better inhibit electron–hole recombination. Zhang et al. [110] proposed a method to dope TiO_2 nanotubes with transition metals, including Fe, Co and Cr, regarding their importance for photoactivity. As presented in Table 3, Fe, Co and Cr are part of the chemical composition of different pigments. Stefanakis et al. [105] synthesized TiO_2 photocatalysts enriched with carbon dots (C-dots) to narrow the semiconductor bandgap.

Furthermore, Sadeghi-Niaraki et al. [99] studied core–shell structures composed of Fe_2O_3 involved with a TiO_2 coating, resulting in a highly reflective colored pigment, which, regarding photocatalytic activity under UV irradiation, even led to the transfer of electrons from the Fe_2O_3 surface to TiO_2. Under visible light, photoactivity could be improved through the transfer of electrons from TiO_2 to Fe_2O_3 [99]. Moreover, an additional intermediate layer of SiO_2 in the composites with Fe_2O_3 and TiO_2 could further increment photocatalytic activity [111]. Adán et al. [112] doped anatase catalysts with different iron contents through a microemulsion method. Therefore, modifying the pigments and their interactions with TiO_2 before adding them to the mortars could eventually enhance the resulting photoactivity.

Libera et al. [113] studied the photoactivity of Fe_xO_y/TiO_2 nanocomposites obtained through atomic layer deposition of iron oxides on the TiO_2 P25 surface. Methylene blue degradation by the nanocomposites was significant even under visible light irradiation, different from what was verified solely using TiO_2 or a mix of Fe_2O_3 and TiO_2 prepared by combining a colloidal solution of the iron oxide with the photocatalyst and further heating at 150 °C for 18 h [113]. On the other hand, pure TiO_2 performed better under UV irradiation than the Fe_xO_y/TiO_2 nanocomposite [113].

Additionally, sensitization of TiO_2 nanoparticles with natural extracts can be suggested as a potential strategy to increase photocatalytic efficiency under visible light irradiation using the injection of electrons with the sensitizing agent into the photocatalyst CB, for instance [114–116].

Although some works [101,102,117] reported the joint study of thermochromic pigments and TiO_2, to the authors' knowledge, the smart pigments were still not mixed with TiO_2-containing mortars, and, thus, there are scientific gaps on the topic, especially regarding the restraints in the thermochromic pigments' durability [117,118]. Moreover, concerning durability, Diamanti et al. [33] exposed photocatalytic colored specimens in Milan, Italy for seven months and observed that brown, red and gray mortars darkened less during winter and easily recovered their initial color, while yellow mortars had similar results as formulations without TiO_2, possibly due to excessive leaching. However, although self-cleaning materials are receiving increasing attention, there is a lack of knowledge about their long-term performance [119]. In addition, there are still difficulties in the photocatalytic evaluation of colored mortars with the degradation of dyes due to the interference of the specimens' original color and the ones of the staining agents [33,55].

Therefore, in summary, photoactivity may observe benefits or losses depending on the type and chemical composition of the pigments and their interaction with TiO_2. There is still a need for further studies on TiO_2-based pigmented mortars, mainly regarding the pursuit of formulations that are able to improve the photoactivity of the materials for use in buildings and enhance their sensitivity to visible light. Modified pigments such as core–shell structures or sensitization processes should be addressed to check the photocatalytic efficiency of the specimens.

Attention should be directed to the durability and environmental impacts of the photocatalytic colored renderings, compared to alternative scenarios, such as painted façades, to ensure and enhance sustainability in the built environment. Indeed, the elimination of the need for painting must be taken into account when assessing the LCA of photocatalytic colored rendering mortars since paints may emit VOCs [120] and result in potentially toxic wastes during their production [121]. Figure 2 summarizes the main findings regarding the interaction of pigments and TiO_2 in mortars.

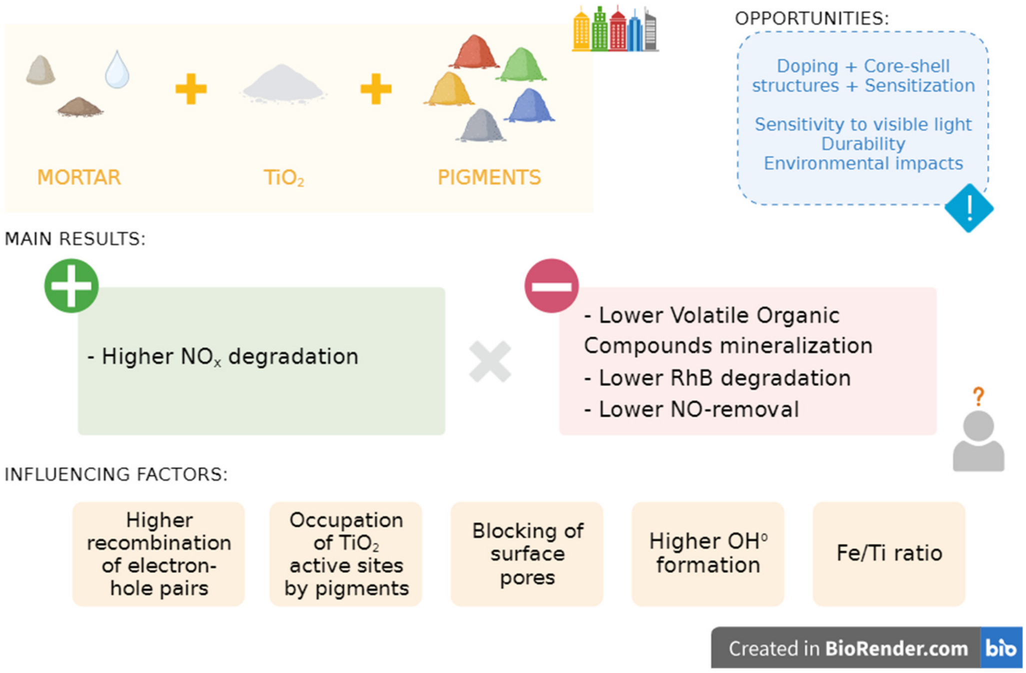

Figure 2. The interaction of pigments and TiO_2 in mortars: main findings and opportunities based on the literature review.

4. Conclusions

This review explored surface finishing characteristics that impact the photocatalytic performance of TiO_2-based mortars in building envelopes. The topic arose from the perspective of elevating the collaboration of TiO_2 in the built environment by enhancing materials' design since the photocatalyst can be an important player in addressing climate concerns by promoting environmental protection, combatting atmospheric pollution, acting against microorganisms, contributing to energy savings and providing self-cleaning effects. All of these further benefit from TiO_2's compatibility with construction materials and the large outdoor area of building envelopes.

The surface roughness and color of the TiO_2-based renderings were mainly addressed. An overview was presented regarding the main findings on the influence of surface roughness on photoactivity, with further emphasis on the evaluation test methods and the surface relationship with water. Regarding colored mortars, an overview of the influence of different types of pigments in non-photocatalytic mortars was developed to substantiate the discussions on their interaction with TiO_2. Final remarks, future trends and opportunities were suggested on the addressed topics.

The surface roughness of TiO_2-based mortars has been investigated concerning both the addition of the photocatalyst to the renderings mix and its application as an additional surface layer. The reported test methods used to evaluate the surface roughness included laser scanning confocal microscopy, atomic force microscopy, profilometers and rugosimeters, among others. Generally, surfaces with moderate roughness performed better regarding photocatalytic efficiency as well as other properties like strength and durability. Microroughness may negatively affect the self-cleaning performance by favoring dirt deposition on the surfaces. TiO_2-based mortars taking advantage of the superhydrophilic effect or addressing superhydrophobicity have been produced, indicating different approaches to obtain the best-performing materials. The importance of an adequate distribution of the photocatalyst over the specimens' surface area is evident. More investigation is needed concerning the influence of surface roughness on the biological growth of photocatalytic mortars. Based on the existing literature, the porosity of the mortars stood out as fundamental for influencing photoactivity.

Concerning TiO_2-based colored mortars, mainly studies with the separate addition of both pigments and the photocatalyst in bulk to the mixture were retrieved from the literature review. Furthermore, the majority of the investigations worked with iron oxide pigments. The effects on photocatalytic efficiency due to the presence of the pigments varied according to their colors and added amounts. Losses in photoactivity were attributed to electron–hole recombination and the pigments' physical occupation of the TiO_2 active sites; increments in photocatalysis were connected to a higher formation of reactive radicals. An opportunity exists regarding modifying and enhancing the pigments with TiO_2 nanoparticles, mainly seeking their improved activation under visible light.

Therefore, regarding surface characteristics of TiO_2-based photocatalytic mortars for use as renderings in building envelopes, their roughness and color impact the final depolluting and self-cleaning performances further than esthetic and functional parameters. Concerning their potential to improve air quality by degrading pollutants and reducing efforts and costs with maintenance actions, further research is suggested approaching the current climate change scenario. Since life cycle information was not identified during this investigation, studies on the topic are strongly recommended as the in-depth experimental test on TiO_2-based mortars' durability. The potential of controlling surface roughness and color on TiO_2-based mortars may be emphasized, seeking better-performing design alternatives which suitably address environmental and functional requirements and ensure sustainability.

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