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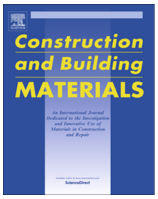

Long term self-cleaning and photocatalytic performance of anatase added mortars exposed to the urban environment

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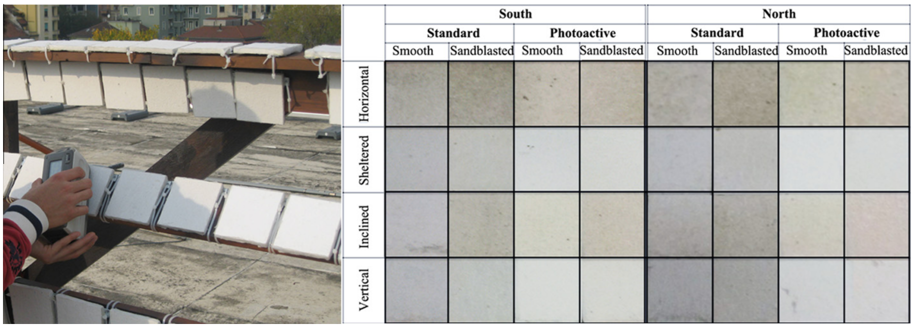
HIGHLIGHTS

- A 2-year natural exposure campaign of photoactive and standard mortars is proposed.
- Lightness and solar reflectance are better maintained if mortars contain TiO_2.
- Aging decreased the materials photoactivity due to soiling.
- Accelerated cleaning with UV and light rinsing restored 70% of initial photoactivity.
- Self-cleaning materials contribute to preserve aesthetics and mitigate heating.

GRAPHICAL ABSTRACT



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ABSTRACT

Building envelope materials containing titanium dioxide have been proposed to exploit their photoactivated depolluting and self-cleaning potential, but a full appraisal of their durability and long-term performance is still missing. This study reports a two-year campaign of natural exposure in Milano, Italy, of photoactive and non-photoactive fiber-reinforced mortars, analyzing the evolution of lightness, solar reflectance, porosity and photoactivity. After aging, photoactive samples showed limited color variation. The photocatalytic activity of TiO_2 containing samples, characterized with dye degradation tests, was minimal after aging. Then, after alternated cycles of UV–Vis irradiation and rinsing, almost 70% of the initial photocatalytic efficiency was recovered.

1. Introduction

The use of TiO_2-modified building materials has been constantly expanding in the last decade, especially in European countries, to exploit their photoactivated depolluting and self-cleaning properties [1–4]. This diffusion is also driven by a growing need for building envelope materials with high solar reflectance and thermal emittance [5–8], or retro-reflective materials that applied onto façades could reflect the solar direct radiation towards the sky, and not towards other buildings [9,10]. In fact, these materials could help to preserve the aesthetics of the building skin, reduce and reshape the energy needs and indoor comfort conditions of buildings [11–14], also contributing to the mitigation of urban

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microclimates [15]. Consider for instance the cooling loads: for the same building, within urban areas the cooling need is in average 13% more than outside the city [16]. This, may yield to an increase by 7% of CO_2 equivalent annual emissions, computed for a reference building in Northern Italy [17]. However, the possible cooling savings and mitigation potential may be compromised by aging [18,19], which is due to the combined action of weathering, soiling, biological growth, and mechanical stress [20–23]. In addition, cleaning techniques do not seem effective to restore the initial reflectance of porous materials such as roofing tiles [24].

The major cause of staining and color variation of building surfaces, reducing the initial solar reflectance, is the accumulation of soot, mainly originated from atmospheric aerosol pollutants such as nitric oxides, carbon based substances and volatile organic compounds [25,26]. Such substances can dissolve in water (i.e., rain and surface condensation) and/or penetrate inside the pores of façade materials (e.g., bricks, claddings, mortars), affecting the aesthetics and reflectance of the façade, and contributing to the physical degradation of external surfaces [27–29].

In this respect, self-cleaning and photocatalytic materials have the added value of a potential prolonged maintaining of their optical performance in spite of soot and particulate matter deposition [30,31], and of mitigating atmospheric pollution [32–35]. The principle on which photoactive materials rely is the activation of a semiconductor through energy provided by light of different wavelength depending on the semiconductor bandgap, generally in the range of near UV or blue visible light. This generates electron/hole couples across the semiconductor bandgap – which in turn induce the formation of highly reactive species, among which hydroxyl radicals play a vital role [36,37]. In fact, these species are then responsible for redox reactions that degrade inorganic and organic compounds adsorbed on the material surface – e.g., volatile organic compounds (VOCs) or NO_x present in the atmosphere. On the other side, the adsorption of the same hydroxyl radicals forms a hydroxylated surface layer that increases hydrophilicity [36,38–40]. The combination of these two mechanisms leads to a self-cleaning effect, where the former helps degrading functional groups by which pollutants adhere to a surface, while the latter spreads water homogeneously over the surface, carrying away particulate matter and degraded contaminants [41–43].

During the last years, many studies have investigated the photocatalytic activity of TiO_2 applied on different materials, in the field of new construction technologies and for cultural heritage preservation [44–47]. Yet, although TiO_2-functionalized building materials are already commercially available, a full appraisal of their long-term performance in use conditions is still missing. Only a few studies in the literature go beyond the measurement of the photodegradation of a given pollutant, or their self-cleaning efficiency in laboratory conditions, and actually propose a long-term approach to this issue [27,40,48–50].

Literature data show that, after aging, the ability of TiO_2 coatings to remove NO_x from air and their self-cleaning ability decreased compared with the initial performance [48]. The loss of TiO_2 efficiency was associated to natural aging after outdoor exposure, especially in the case of coatings subjected to climatic conditions [50,51]. Environmental stress may cause particles detachment and thickness reduction of the coating, owing to the degradation of the coating binder and consequent detachments, as well as a partial deactivation due to the adsorption of pollutants or reaction products of the photocatalytic processes [48,52].

This study reports a two-year campaign of natural exposure in Milano, Italy, of photoactive and non-photoactive fiber-reinforced mortars with different surface finishing, analyzing the evolution of lightness, solar reflectance, porosity and photoactivity of materials.

2. Experimental

2.1. Materials

The materials tested in this work are commercial fiber-reinforced mortars, which are used for rain-screen façade panels as well as pre-cast thermally insulated panels, for new constructions and refurbishment interventions.

For the tests performed in this work, samples composition is reported in Table 1; all mortars were cast with a cement:sand:water ratio of 1:2:0.56. A first fast stirring step was performed to mix water, cement, pigments and chemical additives, followed by a slower mixing where sand and glass fibers were added. The mixture was then extruded on a continuous polystyrene sheet with 8 mm of mortar thickness. Curing in a controlled temperature (25 °C) and relative humidity (65%) chamber lasted 24 h, after which the fiber-reinforced mortar was cut in 100 mm × 100 mm samples, and eventually surface finished if required. Samples with both standard composition and the addition of anatase (a mixture of 2% aqueous suspension and 3% nanopowder, optimized in previous works [53]) were used. Tests were performed on mortars with two different surface finishing conditions, sandblasted and smooth, in order to evaluate the effect of different surface roughness on the self-cleaning performance. XRD (X-ray diffraction) analyses were carried out on the materials used in order to examine the composition of the mortars under investigation.

2.2. Outdoor natural exposure

The selected samples were exposed to the urban environment, on a rooftop of Politecnico di Milano – approx. height 25 m, unsheltered – for a period of two years starting October 2012, in correspondence with the winter activation of buildings heating systems.

Samples were positioned facing both north and south with multiple inclinations (vertical, horizontal, tilted by 45°, and vertical-sheltered) to have a wider understanding of the influence of different microclimates (irradiation, wind, rain), which is also connected with the wetting extent during rain events and therefore with the onset of the superhydrophilicity and self-cleaning (Fig. 1). Samples were sealed with silicone on the four edges and on the back to make them waterproof, and fastened to the racks.

All samples were labeled with reference to their characteristics:

- Sample composition (S: standard, T: with TiO_2).
- Finishing (L: smooth, S: sandblasted).
- Exposure orientation (N: north, S: south).
- Inclination: (H: horizontal, S: vertical sheltered, I: inclined by 45°, V: vertical unsheltered).

Three replicates were originally exposed for each combination of these conditions, for a total of 96 samples. After the first year, one sample per type was withdrawn to perform accelerated photocatalysis tests; consequently the exposure continued with two specimens per type.

Table 1
Composition of mortar samples used in the experimental work.

Composition	Dosage	
Portland cement Roccabianca 42.5R	555 kg/m^3	
Silica sand	1110 kg/m^3	
Water	311 kg/m^3	
Expansive agent Stabilmac	33 kg/m^3	6%
Waterproof additive	22 kg/m^3	4%
Glass fibers	20 kg/m^3	3.6%
Antifoam agent	1 kg/m^3	0.1%