

Photocatalytic nano-composite architectural lime mortar for degradation of urban pollutants under solar and visible (interior) light



Manfredi Saeli^{a,*}, David M. Tobaldi^b, Nejc Rozman^c, Andrijana Sever Škapin^c, João A. Labrincha^b, Robert C. Pullar^{b,*}

^a Department of Architecture (D'ARCH), Polytechnic School, University of Palermo, Viale delle Scienze, Ed. 8, 90128 Palermo, Italy

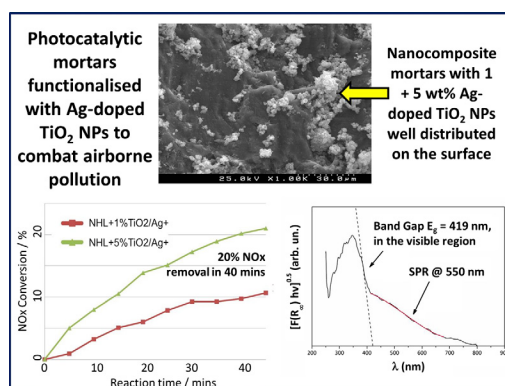
^b Department of Materials and Ceramic Engineering/CICECO–Aveiro Institute of Materials, University of Aveiro, Campus Universitário de Santiago, 3810-193 Aveiro, Portugal

^c Slovenian National Building and Civil Engineering Institute, Dimičeva 12, SI-1000 Ljubljana, Slovenia

HIGHLIGHTS

- Novel functionalised photocatalytic mortar-based nanocomposites are reported.
- Commercial mortar with 1 and 5 wt% Ag-doped TiO₂ (1 mol% Ag) nanoparticle additives.
- Silver doping shifts band gap of titania NPs into visible light wavelengths.
- Nano-additives have no significant effects on curing or colour or mortar.
- Photocatalyst mortars: remove NO_x min under sunlight, VOC removal under white light.

GRAPHICAL ABSTRACT



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ABSTRACT

Recent advances in nano-technology and nano-additives can give enhanced properties to natural hydraulic lime (NHL), creating a multifunctional material. We have prepared a novel nanocomposite, made of a commercial mortar with 1 wt% and 5 wt% added titania nanoparticles (NPs). These TiO₂ NPs are themselves doped with 1 mol% silver, to give the material enhanced photocatalytic and antimicrobial properties. The Ag-doped TiO₂ NPs were made from a simple, costs effective, aqueous green nanosynthesis process, and the end material only contains 0.01–0.05% Ag. As this mortar is intended to both combat atmospheric pollution, and create more durable/lower maintenance building façades (plastering and finishing) by limiting attack from microorganisms and pollutants, its photocatalytic anti-pollution activity under solar and visible (no UV) light for nitrous oxide (NO_x) and volatile organic compound (VOC) removal was studied. The addition of dopants did not significantly alter the physical properties or curing of the mortar, while it showed excellent photocatalytic activity under sunlight. The mortars with only 1 and 5 wt% additives degraded 10.6% and 21% NO_x after 45 mins, respectively, comparing well to a value of 50% for a pure TiO₂/Ag⁺ sample. They also degraded VOCs under visible light, of the kind used for lighting inside buildings, with no UV component. As the NPs are contained in the whole of the mortar layer, not just as a coating, if the surface is chipped or damaged this mortar will not lose its photocatalytic capabilities.

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* Corresponding authors at: Department of Architecture (D'ARCH), Polytechnic School, University of Palermo, Viale delle Scienze, Ed. 8, 90128 Palermo, Italy (M. Saeli), Department of Materials and Ceramic Engineering/CICECO–Aveiro Institute of Materials, University of Aveiro, Campus Universitário de Santiago, 3810-193 Aveiro, Portugal (R.C. Pullar).

E-mail addresses: ing.arch.saeli@hotmail.it, manfredi.saeli@gmail.com (M. Saeli), rpullar@ua.pt (R.C. Pullar).

1. Introduction

The employment of nanomaterials in construction is currently a major research theme worldwide to improve the level of building sustainability (green buildings). Nanotechnology has been demonstrated to be able to enhance the performance of traditional construction materials, producing – by means of standard synthesis procedures – new eco-sustainable, cheap and multifunctional nanocomposites. Nowadays, air pollution is a major concern worldwide as poor air quality – nitrogen oxides (NO_x), volatile organic compounds (VOC) or other micro/nano-particles dispersed in the atmosphere and urban environments – are mainly responsible for an increased risk of respiratory diseases, and may compromise lung function [1–4]. This is true for atmospheres both outside (exterior) and inside (interior) buildings, with a recent increase in “sick building syndrome” due to the design of, and materials used in, modern construction.

Natural hydraulic lime (NHL) is one of the oldest traditional building materials, commonly used in inhabited environments and restoration practice. For centuries, this simple material has undergone few modifications in composition and usage. Lime-based materials are largely used in our living environments (plastering, finishing, masonry binding, etc.), often in addition to cement-based compounds. However, the use of cement is greatly discouraged in preservation and restoration practice of historical architectures, according to European restoration regulations and the numerous recommendations made by dedicated Italian institutions (CNR, ICR, etc.) and international academies, due to the irreparable damage it could cause to the stone matrix [5–7]. The search for better-performing construction and building materials, especially for restoration purposes, is a never-ending pursuit for researchers and manufacturers around the world [8–10]. Thus, research into NHL underwent a recent boost worldwide, especially in the area of the preservation of cultural heritage.

Advances in photocatalysis science and technology, combined with nano-science and nano-technology, make a new generation of enhanced building products possible, with added heterogeneous photocatalysis functionality, and their popularity has grown rapidly over the last decade [11–14]. In addition, beyond the promise of a long-lasting whiteness and cleanness that guarantees less and more-efficient maintenance, photocatalytic construction materials have also been adopted as a promising and effective means to mitigate air pollution.

A large volume of work has been devoted to the research of titanium dioxide (TiO_2) based photocatalysis, and the mechanisms underlying TiO_2 photocatalysis are well known [15–18]. Therefore, at present TiO_2 is the preferred photocatalyst additive for cement/lime-based building/construction materials [19–23]. From an economic perspective, it could significantly reduce operation and maintenance costs, and only solar light is required as a driving force to realise the photocatalytic reaction. TiO_2 is characterised by the high stability of its chemical structure, its biocompatibility, and useful physical, optical and electrical properties. Furthermore, it is widely used in daily real-life applications: as white paint pigment, and in cosmetics, washing/cleaning products, toiletries, food, pharmaceuticals, electrochemical electrodes, solar cells, etc. [24–26]. In recent decades, the interest in TiO_2 has increased for its use as a photocatalytic material, able to oxidise many organic and inorganic compounds [15,27,28]. Indeed, the potential and fascinating applications of heterogeneous photocatalysis with TiO_2 in construction are widespread. Not only in the decontamination and disinfection of polluted waters [29], but also for outdoor and indoor air treatments – indoors, this could be used in order to reduce contaminants that produce the so-called “sick building syndrome”, due to the presence of volatile organic compounds

(VOCs), in interior environments [30,31]. In this way, both outdoor and indoor pollution could be reduced, by a so-called “passive” utilization of multifunctional building materials [19,32]. Furthermore, the production of photocatalytically active construction materials containing titania could enable antimicrobial and self-cleaning surfaces, that might degrade/deactivate organic and microbiological contaminants on their shells, as well as in the surrounding environment, with consequent significant advantageous effects on the health of any occupants/visitors.

A possible way of extending the photocatalytic activity of titania into the visible region is doping it with transition or noble metals [33–35]. More particularly, noble metal modification improves the photocatalytic activity of titania by accelerating liquid–solid and air–solid photocatalytic reactions [36], and by reducing the fast recombination of the photo-generated e^- – h^+ charge carriers [37]. Amongst the noble metals, silver (Ag^+) is particularly attractive due to its remarkable catalytic activity [38], its size, as well as shape-dependent optical properties [39,40], its possible applications in biological and chemical sensing, due to its localised surface plasmon resonance [41], and also because of its long-established and well-known antibacterial properties due to the “oligodynamic effect” [42].

In this work, 1 mol% Ag^+ doped titania was synthesised via an aqueous “green” sol–gel nanosynthesis technique, and successfully combined with natural hydraulic lime mortar. The physical/chemical composition and properties of the specimens were investigated. Their photocatalytic activity was assessed in gas phase, by monitoring the degradation of NO_x under solar lamp radiation, and VOCs under artificial visible light (no UV), showing good results in terms of pollutants removal. The Ag -doped titania permeates the whole of the mortar layer, not just as a coating, so if it is chipped, damaged, cracked or degraded, there will be a fresh surface layer of active NPs ready to continue the anti-pollution functionality.

2. Experimental

2.1. Materials and sample preparation

In order to prepare the silver modified nano-titania powder, a sol–gel synthesis procedure, previously described by Tobaldi et al. [33,35,43,44], was carefully followed to produce 1 mol% Ag^+ doped TiO_2 nanoparticles. The dried gel was thermally treated at 450 °C in a static air flow furnace, heating rate from room temperature to the desired temperature at 5 °C min^{-1} , followed by 2 h soaking time, with natural cooling. Afterwards, the additive powder was ground in an agate mortar to remove agglomeration, and subsequently added to a dry mortar mixture. These silver- TiO_2 NPs are composed of 85.5 wt% anatase, 5.2 wt% rutile and 8.3 wt% brookite phases of titania, and the average diameter of the anatase NPs is 7.8 nm [45]. The pure NPs have been well characterised (XRD, TEM, optical and photocatalytic properties) in references [33,35,43,44].

Natural hydraulic lime mortar (Weber Rev 158, supplied by Saint Gobain-Weber, Portugal) was used as a binding material. In this study, to prepare the mortar specimens, the water/lime ratio was 0.28 (280 g water to 1000 g NHL, as recommended by the company). At this point, 1 wt% and 5 wt% of the silver-modified nano-titania powder (Ag-TiO_2) was also added to the NHL mortar (e.g., 10 g Ag-TiO_2 to 1000 g NHL for a 1 wt% mix), in order to study the effects of such different quantities added to the mixture.

The procedure for the preparation of the specimens was standardised following the UNI EN 1015–2:2007 procedure. Firstly, all the proportioned materials (lime/additives/distilled water) were mixed uniformly for about 5 min using a mechanical mixer. Then, specimens were moulded in cylinders (22 mm diameter, 10 mm height) and cured at a temperature of 20 ± 2 °C for 28 days, at a relative humidity (RH) = $95 \pm 5\%$ for 7 days, then RH = $65 \pm 5\%$ for the last 21 days. At the end of the curing period, specimens were physically and chemically characterised, in particular as regards their photocatalytic activity.

2.2. Characterisation

Diffuse reflectance spectroscopy (DRS) was performed with a Shimadzu UV 3100 spectrometer. Spectra of the specimens were acquired in the UV–vis range (250–800 nm), with 0.2 nm step size and with the use of an integrating sphere and a white reference material, both made of BaSO_4 .

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