ARTICLE IN PRESS

Science of the Total Environment xxx (2016) xxx-xxx

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv



Environmental and human health assessment of life cycle of nanoTiO₂ functionalized porcelain stoneware tile

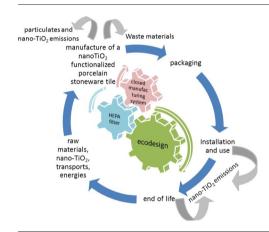
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HIGHLIGHTS

- A preliminary frameworks to evaluate human toxicity and exposure factors for nanoparticle releases was implemented.
- Ecodesign of an industrial scale up of a nanoTiO₂ functionalized glazed stoneware tile.
- Precautionary actions were defined in order to reduce the potential toxicity of nanoparticles in the entire life cycle.
- The most critical environmental burdens and the benefits related to the use of nanoTiO₂ were assessed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 24 May 2016
Received in revised form 9 October 2016
Accepted 16 October 2016
Available online xxxx

Editor: Simon Pollard

Keywords: LCA nanoTiO₂ Functionalized ceramic tile Eco-design Human toxicity

ABSTRACT

Recently, there has been a rise in the interest in nanotechnology due to its enormous potential for the development of new products and applications with higher performance and new functionalities. However, while nanotechnology might revolutionize a number of industrial and consumer sectors, there are uncertainties and knowledge gaps regarding toxicological effects of this emerging science. The goal of this research concerns the implementation into Life Cycle Assessment (LCA) of preliminary frameworks developed to evaluate human toxicity and exposure factors related to the potential nanoparticle releases that could occur during the life cycle steps of a functionalized building material. The present LCA case study examines the ecodesign of nanoTiO₂ functionalized porcelain stoneware tile production. The aim of this investigation is to manufacture new eco-friendly products in order to protect human health and ecosystem quality and to offer the market, materials with higher technological properties obtained by the addition of specific nanomaterials.

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1. Introduction

In recent years, a huge amount of researches have been conducted for the development of innovative processes and eco-friendly

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technologies based on photocatalytic properties of solid semiconductors that are able to oxidize organic and inorganic pollutants until their complete mineralization (Ibhadon and Fitzpatrick, 2013).

In the field of photocatalytic construction and building materials, titanium dioxide is the most widely used photocatalyst showing greater photocatality activity in the crystal structure of anatase (Quagliarini et al., 2013; Chen and Poon, 2009). In particular, the

http://dx.doi.org/10.1016/j.scitotenv.2016.10.115 0048-9697/© 2016 Elsevier B.V. All rights reserved.

Please cite this article as: Pini, M., et al., Environmental and human health assessment of life cycle of nanoTiO2 functionalized porcelain stoneware tile, Sci Total Environ (2016), http://dx.doi.org/10.1016/j.scitotenv.2016.10.115

photocatalytic properties of anatase improve when this material is used in the form of nanocrystals, with characteristic dimension in the order of nanometers (10^{-9} m). The nanometric dimensions are responsible for the high ratio between surface area and volume, with consequent increase of surface sites where photocatalytic activity takes place. In this way, when exposed to radiation of adequate wavelength (388 nm), nanotitanium dioxide (TiO_2) shows peculiar characteristics like photocatalysis of redox reactions, but also superhydrophilicity and antibacterical properties. The functionalized surface has an improved cleanability and antibacterial activity but it also becomes active, i.e., to reduce the concentrations of air pollutants such as nitrogen oxides (NO_x) and volatile organic compounds (VOC_s) deposited on or in contact with the material surface.

Nanotitania can functionalize different types of surfaces, such as metals, glass and resins, composing architectural and decorative indoor and outdoor elements, to obtain specific surface properties. TiO₂ nanoparticles surfaces can find applications in private and public buildings (i.e. schools, hospitals, car parks, tunnels) especially in closely bacterial monitored structures, such as operating theatres, swimming pools and sports fields. For example, functionalized self-cleaning and anti-smog steel slabs can be used as coating of exterior ventilated walls or in screening systems such as sunshade slats (Marinelli et al., 2013). NanoTiO₂ coated float glass has been considered for applications in private buildings for traditional windows and curtain walls, as well as glazings to create self-cleaning and anti-fogging surfaces and to ensure heat gain during winter and summer seasons (Pini et al., 2013). Aluminium panel coated with polyurea resin functionalized with nanoTiO2 has been studied for both indoor applications with decorative and self-sterilizing functions and outdoor applications as self-cleaning, air depollution and anti-fogging coating (Pini et al., 2012).

The development of new easy-to-clean or even self-cleaning surfaces has recently also been the main research field for ceramic tile industries that are investigating different surface structures using nanoparticles and nanocoatings (Berto, 2007). In fact, among the different and interesting properties of nanomaterials, they are characterized by a mean diameter below the light wavelength and thus they are transparent if applied on a substrate or dispersed in a matrix. This aspect is particularly important for materials, such as ceramic tiles, for which the aesthetic aspect is often the parameter that determines the choice or the impression one has of them, which are rarely determined by the particular functional properties. In the last five years, a large number of tile ceramic factories are trying worldwide to overcome some of the technological challenges in order to scale up the process and obtain a large-scale application. In particular, the main problem is represented by the need to avoid the anatase to rutile phase transformation, which implies a reduction of photoactivity but also excludes the application of the photocatalyzer before the sintering step that is performed industrially at 1200-1220 °C (Raimondo et al., 2012). Several strategies have been studied to overcome these problems. As an example, Bondioli et al. suggested the use of sol-gel coating (Bondioli et al., 2009; Niederhausern et al., 2013) while Szczawiński et al. (2011) RF diode sputtering, atmospheric pressure chemical vapour deposition (APCVD) and spray pyrolysis deposition (SPD).

The growing employment of titania nanoparticles in building materials has increased the probability of their accidental or incidental release into the environment and, thereby, of possible human exposure at different stages of their life cycle.

Life cycle assessment (LCA) is the most adequate methodology to assess the environmental and health effects of a product, process and service. Moreover, it has been recognized as a powerful "tool" to assess the environmental performance of nanoproducts.

Over the last years numerous review articles about the use of LCA in the nanotechnology field have been published (Hischier and Walser, 2012; Gavankar et al., 2012; Upadhyayula et al., 2012; Kim and Fthenakis, 2013; Lazarevic and Finnveden, 2013; Miseljic and Olsen, 2014). Hischier et al. (2016) examined these works and ended up that

three main issues should be taken into account in order to proper model LCA of nano-enabled application:

- 1. the use of an adequate functional unit, taking into account any additional functionality that the use of nanotechnology brings;
- the lack of reliable inventory datasets about the release of nanomaterials into the environmental compartments;
- 3. the urgency of developing appropriate characterization factors (CFs) for nanomaterials, for both humans and the environment.

The authors deem indispensable to cover the third issue in order to be able to actually assess the life cycle of nanoproducts in the life cycle impact assessment (LCIA) step.

To cope with this problem this study implemented the preliminary human health toxicity factors for nanoTiO₂ developed following Ecoindicator 99 procedure (Ferrari et al., 2015 and Pini, 2015) and USEtox™ method (Pini et al., 2016) into LCIA stage.

The here proposed LCA study concerns the ecodesign of an industrial scale up of a nanoTiO₂ functionalized glazed porcelainized stoneware tile. The porcelainized stoneware tile, also called grés, is a product used both for internal and external applications in building field. Lowporosity, high mechanical, abrasion, chemical and stain resistance make this material ideal to flooring areas with elevated public use. Its high technical properties are mainly due to an extremely sintered body composed of different crystalline phases (anorthite, mullite and quartz) immersed in a vitreous matrix. In the ceramic tiles industrial field, the porcelainized tiles have become more and more important with regard to its spread from very few market shares limited as to their application fields to more and more diversified ones; the result has been a clear increase in production volumes. This product, which was formerly considered only from a technical standpoint, nowadays shows high aesthetical potentialities allowing its use for over-refined purposes. The possibility to obtain a functionalized tile surface, that becomes active thanks to the titania nano particles towards pollutants and bacteria, is a strong challenge both from a scientific and industrial point of view (Sciancalepore and Bondioli, 2015).

Because of the limited knowledge currently available regarding the effects nanoTiO₂ may have on environment or human health (lavicoli et al., 2012), a safe behavior has been adopted in all life cycle steps where workers could come into contact with or inhale nanoparticles released by nanocoating surface. Nanoparticles releases have been assessed during all involved life cycle stages (e.g. application, handling, installation, use phase and end of life treatment). In agreement with the ecodesign approach, the production choices adopted in the present study have been finalized to minimize the environmental and human health impacts. Moreover, the implementation of these new the preliminary human health toxicity factors for nanoTiO₂ in the impact assessment phase help to better understand the most critical environmental and human burdens associated to the releases of nanoparticles during the entire life cycle of this innovative building nanomaterial.

In this work, the patent procedure for the preparation of aqueous suspension of nanoTiO $_2$ has been adopted to model the nanoparticle synthesis (Colorobbia, 2014) and some environmental benefits derived from the photocatalytic effect of nanoTiO $_2$ have been considered, e.g. the reduction of organic and inorganic emission concentrations and the reduction of the use of cleaning agents. At the time of LCA modelling the latest and available version of Ecoinvent database was v2.

Recently, even Hischier et al. applied into LCA case study using nanoTiO₂ in outdoor façade coatings (Hischier et al., 2016) human toxicity factors but this time only CFs performed by USEtox™ method (Pini et al., 2016). The scope of Hischier et al.'s work is to investigate the influence of variations on the three main above mentioned challenges regarding LCA and nanotechnology field. In order to achieve this goal, they examined >200 different scenarios, which took into account: *i*) different study set up such as the influence of a change of the actual lifetime of the nanoTiO₂ coating paint and change in the formulation; *ii*) different nanoTiO₂ synthesis routes (Hischier et al., 2015; Pini et al.,

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