

Water based green sol-gel synthesis of TiO₂ sol for photocatalytic water and air purification coatings with self-cleaning nature

M. Ayyaz,^{a,b} B. Aksu,^a M. J. Mateos,^b M.P. Pedferri,^a M.V. Diamanti,^a S. Suarez^b

^a Department of Chemistry, Materials and Chemical Engineering “Giulio Natta”, Politecnico di Milano, Via Mancinelli 7, 20131 Milan, Italy.

^b FOTOAIR-CIEMAT, Energy and Environmental Devices Unit, Energy Department, Avda. Complutense 40, Madrid, Spain.

Environmental pollution is a pressing concern of modern society, as air, water and soil contamination caused by progressing industrial societies are directly affecting the ecosystem. The European Union (EU) is also committed for a sustainable environment by reducing the pollution to safe levels for human health and ecosystem. The EU Green Claims Directive [1] and EU Green Deal vision 2050 [2] raise critical concerns for sustainable science of materials as it strict the verification rules under the EU Green Claims Directive to use economic and eco-friendly processes and protocols for materials synthesis. In pursuit of sustainable environmental remediation technologies, TiO₂ photocatalyst coatings have garnered significant interest of both industrial and researcher community. Numerous methods have been employed by researchers for synthesis of TiO₂ based coatings for environmental remediation. However, most of these methods involve harmful reagents such as organic solvents, strong acid catalysts and some other additives which pose a serious threat to both human health and ecosystem [3].

This research presents the effect of temperature and aging time on green sol-gel synthesis of TiO₂ sol for the preparation of highly uniform and adherent coating. Titanium tetraisopropoxide (TTIP) was used as the precursor, with water as solvent/hydrolysing agent and acetic acid as catalyst/chelating agent serving as eco-friendly reagents under mild conditions. TiO₂ layers were immobilized on glass substrates via dip-coating technique, evaluating their properties for Rhodamine B photooxidation in water and formaldehyde in gas phase under UV-A irradiance. Moreover, the self-cleaning properties were evaluated according to ISO 27448:2009 standard by the photodegradation of oleic acid under UV-A exposure [4]. The results revealed a critical sensitivity of the TiO₂ sol to ambient conditions, which significantly affected sol aging behaviour, quality and photocatalytic activity of coatings. TiO₂ sols synthesized at ambient conditions between 26-32 °C characteristic of summer season in Mediterranean countries and aged progressively, developed sufficient stability and activity after 21 days. DLS revealed formation of nanoparticles (10-100 nm) in these sols with an increasing trend over aging time indicating particles growth and colloidal instability. These sols yielded highly uniform and efficient coatings with excellent photodegradation of Rhodamine B. Conversely, when the synthesis method was performed at temperatures ranged between 15-18 °C during winter season, the resulting sols remained low viscous leading to inefficient coatings with lower amount of TiO₂ (≤ 0.026 mg/cm²). DLS confirms the presence of nano scale particle distributions. Various attempts to get efficient coatings were unsuccessful even by increasing TiO₂ amount and varying layer drying or the annealing temperature.

To overcome this issue, an accelerated aging protocol was opted by incubating the sol at 40 °C for 21 hours. This controlled heating triggered partial oligomerization with, increasing the viscosity of the sol, leading to formation of homogeneous and adherent coatings with 0.106 mg/cm² amount of TiO₂. A single-layer coating of this sol exhibited exceptional degradation of formaldehyde in gas phase (10 ppm) under UV-A irradiance with 100%, 73%, and 56% removal at 300, 500, and 700 mL.min⁻¹ flow rates, respectively. All coatings showed a photoinduced super hydrophilic behaviour with the water contact angle measurements with values below 10° after 180 min under UV-A exposure, confirming effective surface activation and strong self-cleaning nature. These findings underscored the critical connection between aging conditions and sol rheology properties for fabrication of efficient photoactive coatings.

This study emphasizes on temperature-mediated aging as a decisive factor for reproducibility and performance of green sol-gel derived TiO₂ coatings. The proposed strategy of accelerated aging supports a simple and scalable route to prepare TiO₂ based green photocatalytic materials for indoor air purification and other environmental remediation technologies.

References

- [1] EU Commission. (2023). COM/2023/166 final. <https://eur-lex.europa.eu/legal-content/EN/TXT>.
- [2] European Commission. “Towards Zero Pollution for Air.” Water and Soil”-COM 400 (2021): 1-22.
- [3] Chang, C., Rad, S., Gan, L., Li, Z., Dai, J., & Shahab, A. Nanotechnol. Rev., 12 (2023), 20230150.
- [4] Mateos, M. J., A. Lao-Zea, M. P. Alcacer, B. Sánchez, and S. Suárez. J. Photochem. Photobiol. A: Chem. 457 (2024): 115880.

Acknowledgements

This study was carried out within the MICS (Made in Italy —Circular and Sustainable) Extended Partnership and received funding from the European Union Next-Generation EU (PIANO NAZIONALE DI RIPRESA E RESILIENZA (PNRR) MISSIONE 4 COMPONENTE 2, INVESTIMENTO 1.3 - D.D 1551.11-10-2022, PE00000004) and MCIN/AEI Spain (PID2022-138682OB-I00).