**Design and properties of Titanium Dioxide/Graphene Oxide composites exploitable in wastewater treatments**

Simona Ortelli[1], Lara Faccani[1], Enrico Ercolani[1,2], Ilaria Zanoni[1], Chiara Artusi[1], Magda Blosi[1], Stefania Albonetti[2], Anna Luisa Costa[1]

[1] CNR-ISSMC, National Research Council of Italy - Institute of Science, Technology and Sustainability for Ceramics, Via Granarolo 64, 48018 Faenza, Italy

[2] Department of Industrial Chemistry "Toso Montanari", Bologna University, Viale Risorgimento 4, 40136 Bologna, Italy

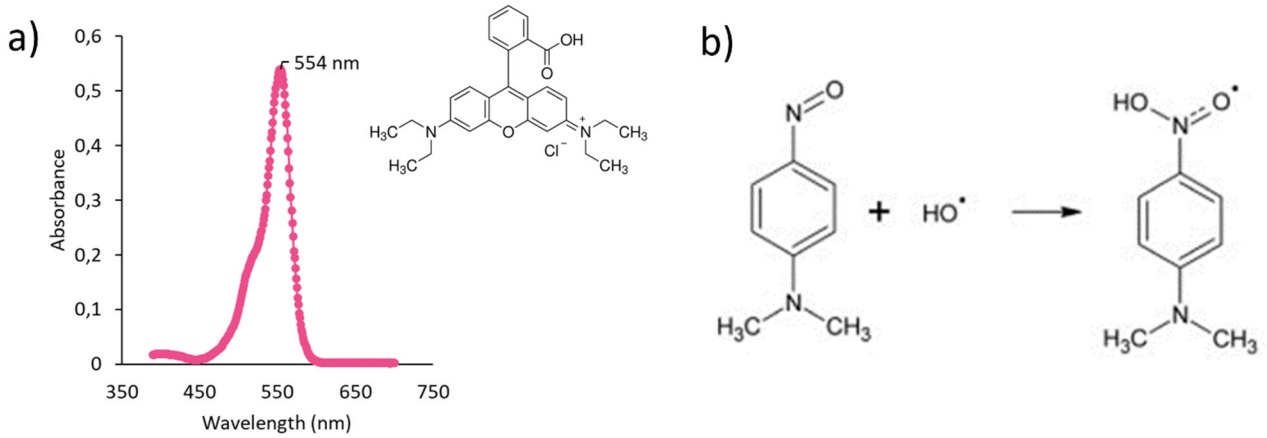



Figure S1. a) Rhodamine B absorbance spectra and molecular strcucture; b) oxidation reaction scheme of RNO.

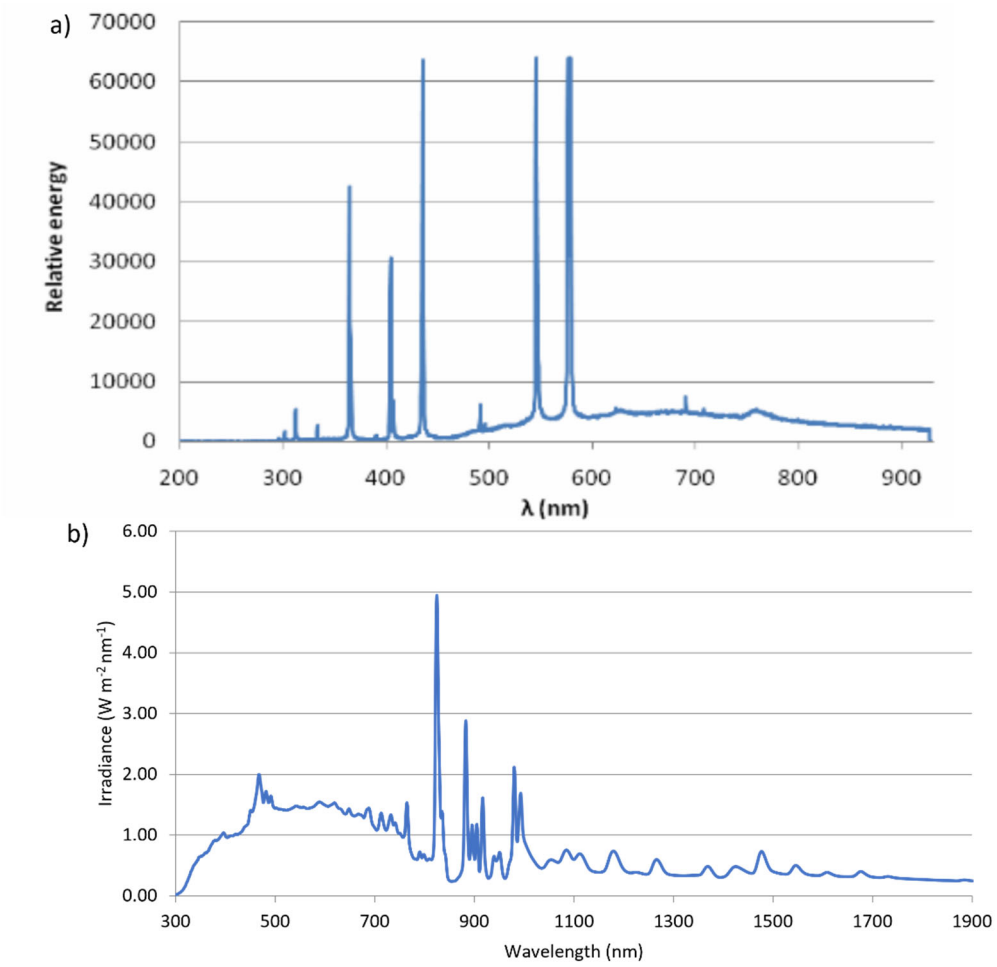
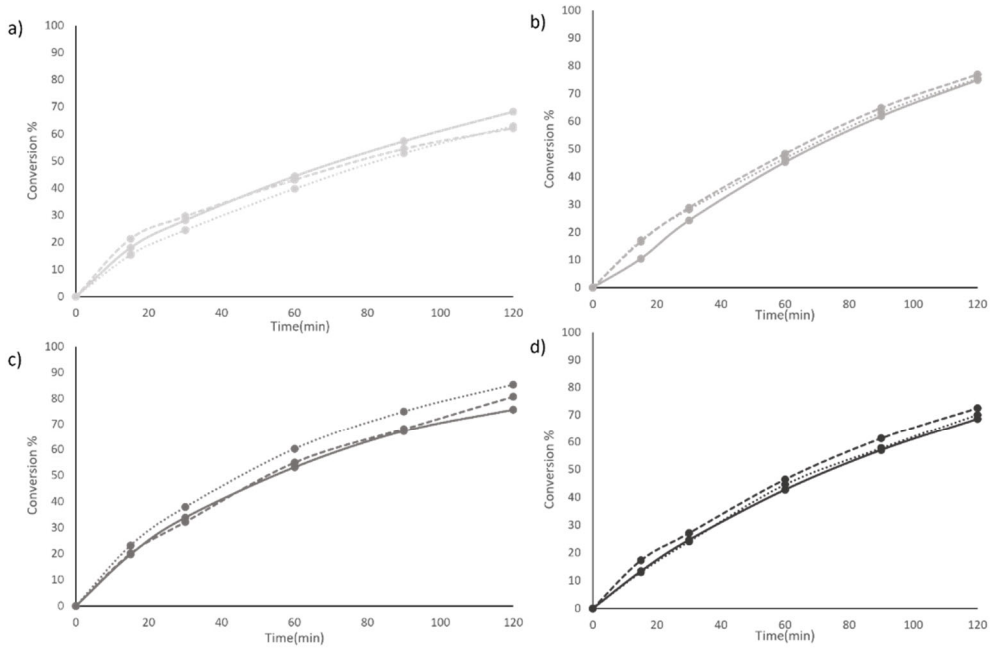
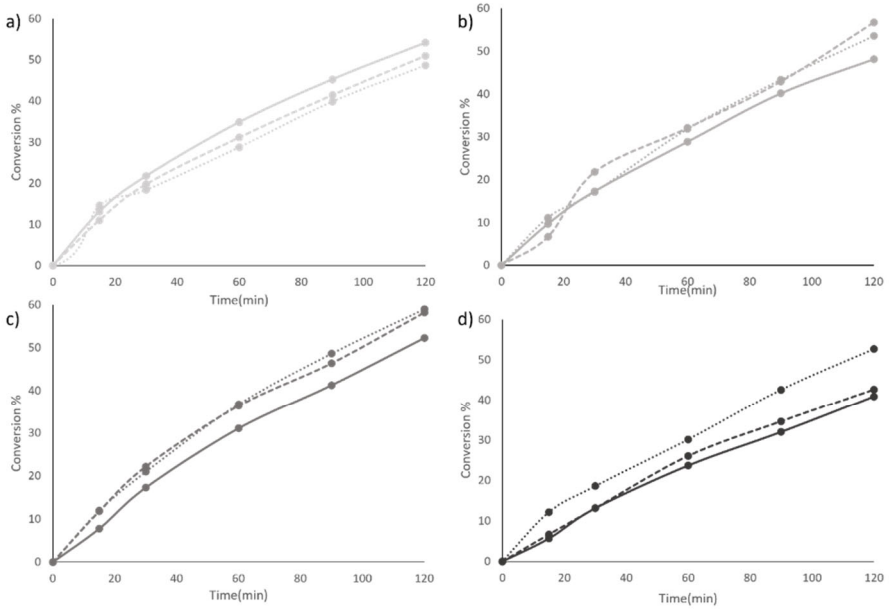



Figure S2. Emission spectrum of a) UV lamp and b) solar simulator.




| time (min) | Standard deviation | | | |
|---|---|---|---|---|
| | $TiO_2$ DT-51 | TGO_9 | TGO_16 | TGO_25 |
| 0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 15 | 3.0 | 3.7 | 1.9 | 2.4 |
| 30 | 2.6 | 2.5 | 2.9 | 1.6 |
| 60 | 2.4 | 1.5 | 3.7 | 1.9 |
| 90 | 2.2 | 1.5 | 4.2 | 2.3 |
| 120 | 3.3 | 1.0 | 4.9 | 2.0 |

Figure S3. Photocatalytic degradation of RhB, under UV light, using as catalyst a) $TiO_2$ DT-51; b) TGO_9%; c) TGO_16% and d) TGO_25% and corresponding standard deviation.




| time (min) | Standard deviation | | | |
|---|---|---|---|---|
| | $TiO_2$ DT-51 | TGO_9 | TGO_16 | TGO_25 |
| 0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 15 | 1.8 | 2.3 | 2.4 | 3.5 |
| 30 | 1.7 | 2.6 | 2.6 | 3.2 |
| 60 | 3.1 | 1.8 | 3.2 | 3.3 |
| 90 | 2.8 | 1.7 | 3.8 | 5.5 |
| 120 | 2.8 | 4.3 | 3.7 | 6.4 |

Figure S4. Photocatalytic degradation of RhB, under solar light, using as catalyst a) $TiO_2$ DT-51; b) TGO_9%; c) TGO_16% and d) TGO_25% and corresponding standard deviation.

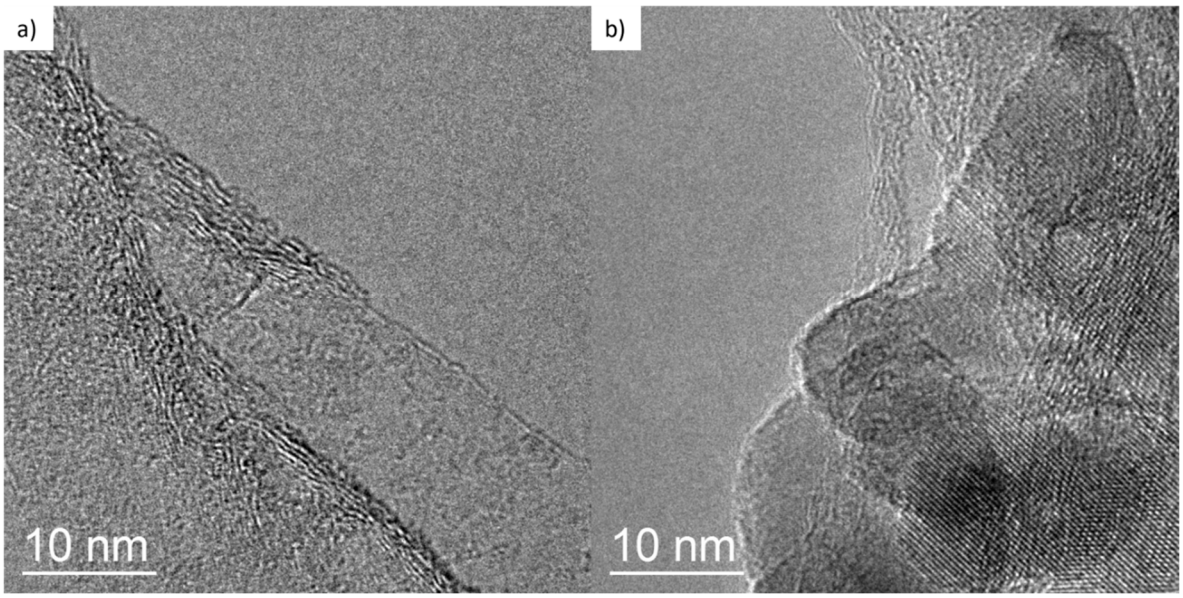



Figure S5. HRTEM images of a) GO and b) TGO_16% sample.

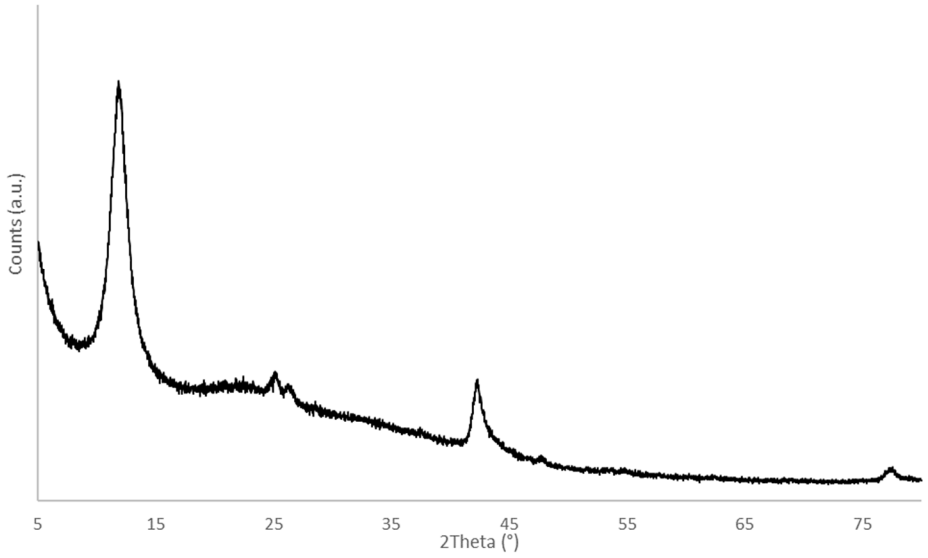



Figure S6. XRD patterns for GO.

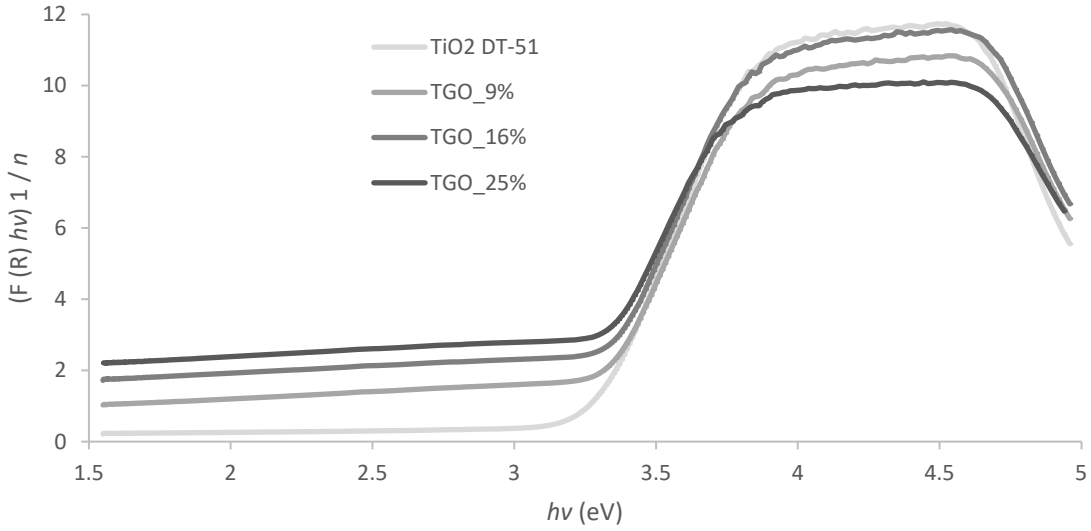



Figure S7. Tauc's curve for $TiO_2$ DT-51 and TGO composites.

Table S1. Specific surface area of $TiO_2$, GO and TGO composites.

| Sample | s.s.a. ($m^2$/g) |
|---|---|
| $TiO_2$ DT51 | 91 |
| GO | 23 |
| TGO_9% | 121 |
| TGO_16% | 76 |
| TGO_25% | 69 |

Table S2. Results of $Cu^{2+}$ adsorption (% $Cu^{2+}$ adsorbed with relative standard deviation) tests

| Sample | Time (min) | | | | | |
|---|---|---|---|---|---|---|
| | 1 | 5 | 15 | 45 | 60 | 1440 (24h) |
| $TiO_2$ DT-51 | 5 ± 2.6 | 32 ± 1.1 | 18 ± 0.8 | 34 ± 0.8 | 36 ± 2.1 | 22 ± 0.4 |
| GO | 69 ± 0.3 | 81 ± 1.4 | 85 ± 0.6 | 79 ± 0.2 | 97 ± 0.9 | 85 ± 1.3 |
| TGO_9% | 25 ± 1.0 | 12 ± 3.0 | 17 ± 0.8 | 22 ± 4.8 | 28 ± 1.1 | 25 ± 2.3 |
| TGO_16% | 26 ± 4.1 | 37 ± 7.3 | 43 ± 4.5 | 52 ± 5.5 | 51 ± 1.0 | 42 ±8.3 |
| TGO_25% | 19 ± 3.3 | 32 ± 2.9 | 39 ± 1.7 | 37 ± 7.3 | 36 ± 1.8 | 33 ± 2.3 |

Table S3. Summary of photocatalytic (RhB conversion %, after 120 min and moles RNO consumed/mg of phase dispersed, after 1 h) and adsorption (% $Cu^{2+}$ adsorbed, after 60 min) results.

| Sample | RhB conversion % | | moles RNO consumed/mg of phase dispersed | Adsorption % $Cu^{2+}$ adsorbed |
|---|---|---|---|---|
| | Under UV light | Under solar light | | |
| $TiO_2$ DT-51 | 64 ± 3.3 | 51 ± 2.8 | 4.3E-07 ± 4.09E-10 | 36 ± 2.1 |
| GO | nd* | nd* | 1.22E-07 ± 7.90E-09 | 97 ± 0.9 |
| TGO_9% | 76 ± 1.0 | 53 ± 4.3 | 3.67E-07 ± 1.08E-9 | 28 ± 1.1 |
| TGO_16% | 80 ± 4.9 | 56 ± 3.7 | 6.20E-07 ± 1.78E-09 | 51 ± 1.0 |
| TGO_25% | 70 ± 2.0 | 45 ± 6.4 | 4.19E-07 ± 2.49E-09 | 36 ± 1.8 |

* *not determined*

Table S4. The adsorption capacity in this study compared with previous studies.

| Sample | Dye or metal | Adsorption capacity | Reference |
|---|---|---|---|
| TGO - 25% | MB | 20.25mg/g | [50] |
| $TiO_2$/GO nanocomposite | $Cd^{2+}$<br>$Pb^{2+}$ | 69,36%<br>89% | [51] |
| GO, $TiO_2$ and $MgFe_2O_4$ nanocomposite | MB | 99% | [52] |
| GO/$TiO_2$ nanocomposite | RhB<br>$Cd^{2+}$<br>$Zn^{2+}$ | 11.299 mg/g<br>14.326 mg/g<br>11.326 mg/g | [53] |
| GO/$ZnTiO_3$/$TiO_2$ | MB | 78 mg/g | [54] |
| $TiO_2$-GO aerogel | $Cu^{2+}$ | 39.8 mg/g | [55] |
| TGO_16% | $Cu^{2+}$ | 52% | Present study |

The hypothetical RhB photodegradation mechanism is shown below

$TiO_2 + hv \rightarrow TiO_2 (e^-) + TiO_2 (h^+)$

$GO + e^- \rightarrow GO(e^-)$

$h^+ + OH^- \rightarrow {}^\bullet OH$

$h^+ + H_2O \rightarrow {}^\bullet OH + H^+$

$e^- + O_2 \rightarrow {}^\bullet O_2^-$

TGO ($^\bullet OH$ or $^\bullet O_2^-$) + RhB $\rightarrow CO_2 + H_2O$