## Exfoliated g-C<sub>3</sub>N<sub>4</sub> for CO<sub>2</sub> conversion into fuels and chemicals

G. Ramis<sup>1,</sup>, M. Tommasi<sup>2</sup>, S. Naz Degerli<sup>3</sup>, I. Rossetti<sup>2,3</sup>

<sup>1</sup> Dip. Ing. Chimica, Civile ed Ambientale, Università degli Studi di Genova and INSTM Unit Genova, Genoa, Italy

<sup>2</sup> Chemical Plants and Industrial Chemistry Group, Dip. Chimica, Università degli Studi di Milano

<sup>3</sup> INSTM Unit Milano-Università, Milan, Italy

## Abstract

With rising concerns on climate change and fossil fuels consumption, significant efforts have been made to reduce CO<sub>2</sub> emissions and produce chemicals/fuels from renewable feedstocks. Solar driven CO<sub>2</sub> photoreduction with advancement in heterogeneous catalysis is promising, albeit challenging due to low quantum efficiency and limited solar light absorption. Layered twodimensional materials like graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) can pave the way considering intriguing properties and practical applications. However, an additional post-synthesis step of exfoliation is needed to enhance surface area and optoelectronic properties of such materials, after their synthesis in bulk form. Herein, we demonstrate exfoliation of graphitic carbon nitride g-C<sub>3</sub>N<sub>4</sub> by means of UltraSound (US) treatment using water as a solvent at varying input power at constant frequency, constant amplitude and time of effective sonication. This can positively contribute to the properties of the final material without critical handling or environmental issues. Exfoliation of g-C<sub>3</sub>N<sub>4</sub> in water displays a strong dependence of US input power, with a slightly enhanced bandgap (2.8 eV), but most of all increased lifetime of photogenerated as observed through Diffuse Reflectance Spectroscopy electrons. (DRS) and Spectrofluorimetry data. Among all applied power (varied between 30W and 120W), 120W sufficiently exfoliated and tuned physicochemical properties of  $q-C_3N_4$ . Compared to bulk as prepared sample, exfoliated g-C<sub>3</sub>N<sub>4</sub> exhibited improvement in photoinduced charge carrier transfer and separation, resulting in higher photocatalytic efficiencies.

Accordingly, the bandgap and charges lifetime of the materials correlate well with change in input power and, as well, the catalytic performance determined through an innovative high-pressure reactor in solid-liquid-gas phase. The catalytic results demonstrate this metal free material as an efficient photocatalyst to obtain high yield of formic acid with productivities ranging from ~5100 to ~8200 mmol/kg<sub>cat</sub> h at 80°C in water, which is among the highest reported in the literature.

The financial contribution of Fondazione Cariplo through the grant 2021-0855 – "SCORE - Solar Energy for Circular  $CO_2$  Photoconversion and Chemicals Regeneration", funded in the frame of the Circular Economy call 2021 is gratefully acknowledged.