ABSTRACT

Emerging contaminants are infiltrating freshwater sources at alarming rates, with levels expected to rise in the coming years. This underscores the urgent need for cutting-edge wastewater treatment processes to safeguard human health and well-being. Notably, heterogeneous photocatalysis stands out as a powerful technology due to its ability to transform a broad spectrum of micropollutants, including inorganic, organic, and biological contaminants, into innocuous species, rather than merely transferring them between environmental compartments. In recent years, two-dimensional (2D) materials, particularly metal-free graphitic carbon nitride (g-C_3N_4), have attracted considerable attention for photocatalytic applications owing to their intriguing physicochemical properties. However, pristine g-C_3N_4 suffers from several noteworthy limitations, including limited visible light absorption and rapid recombination of photogenerated charge carriers. Furthermore, the practical implementation of 2D materials in aqueous photocatalytic systems is hindered by their tendency to restack, which decreases the accessible surface area and reduces catalytic efficiency. Such aggregation can also lead to sedimentation, limiting light penetration and thereby diminishing photocatalytic activity. In addition, due to its powdery nature, incomplete post-treatment recovery of g-C_3N_4 results in catalyst loss and potential secondary pollution. To address these challenges, engineering g-C_3N_4 through doping and heterojunction formation, as well as assembling it into three-dimensional (3D) macrostructures, represents an effective strategy. This is because 3D architectures can foster synergistic interactions among the engineered components, thereby markedly enhancing photocatalytic performance. By assimilating the complementary electronic, optical, and structural attributes of individual materials within a single framework, these 3D macroscale materials enable improved

light harvesting, promote efficient charge separation, and provide a robust platform for convenient catalyst recovery. In this context, the present doctoral thesis explores the photocatalytic performance of 3D macrocomposites of modified g-C_3N_4 with the objective of eliminating diverse classes of micropollutants, including heavy metal ions, pharmaceutical residues, and antibiotic-resistant bacteria, from aqueous environments. The findings of this work advance the practical applicability of g-C_3N_4 in heterogeneous photocatalysis and open new avenues for the efficient removal of emerging contaminants, thereby supporting the development of sustainable wastewater treatment technologies.

Keywords*:* heavy metals, pharmaceuticals, antibiotic-resistant bacteria, heterogeneous photocatalysis, visible light, 2D semiconductors, graphitic carbon nitride, metal oxides, molybdenum disulphide, heterojunction, elemental doping, aerogel, 3D printing, interfacial charge transfer, real water matrices.