Tailoring the stoichiometry of graphitic C$_3$N$_4$ nanosheets under electron beam irradiation

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Figure S-1 (A) Vials containing C$_3$N$_4$ powder on the left and a small amount of C$_3$N$_4$ powder bath sonicated and dispersed in isopropanol on the right. After the sonication, the solution containing the dispersed flakes were left resting for 600 s to promote the precipitation of large agglomerations. Only the supernatant was used to drop coat the TEM grid. The table summarizes the parameters used to prepare the TEM sample. The various sonication times to disperse and exfoliate the C$_3$N$_4$ flakes are shown in panels (B)-(E). The C$_3$N$_4$ flakes are shaded in green. Panel C shows shaded in red an amorphous carbon Lacey film supporting the C$_3$N$_4$ flakes. No differences in the exfoliation efficiency between the shortest and longest sonication times were observed. To minimize defects short sonication times were used.
Figure S-2 (A)-(A'') C₃N₄ powder bath sonicated for 180 s in isopropanol and imaged at 300 kV. The flakes are crystalline and it is possible to acquire high-resolution images with low current densities to minimize electron beam irradiation damage. Applying an FFT filter makes it easier to observe the lattice, as seen in A'. Panel A'' shows the FFT of the initial image A. (B) to (B'') C₃N₄ powder sample bath-sonicated for 1800 s to check if the exfoliation of the C₃N₄ layers is better. The images above show that there is no noticeable difference between sonication times.

Figure S-3 Comparison of the degradation time with respect to the microscope acceleration voltage and the electron current density. (A) and (A') suggest that using an acceleration voltage of 300 kV the degradation of the C₃N₄ lattice happens at a fast rate for current densities above 4.0x10⁻¹¹ nA/nm². The same case is for acceleration voltage of 80 kV shown in panels (B) and (B'). For 60 kV rapid degradation of the lattice occurs for current densities above 3.0x10⁻¹¹ nA/nm² shown in panels (C) and (C'). For 30 kV the degradation of the lattice occurs at lower current densities, typically above 2.0x10⁻¹¹ nA/nm² as shown in panels (D) and (D').