Optimizing Cobalt Loaded C_3N_4 Photocatalysts via Heat Exfoliation for Reducing CO_2

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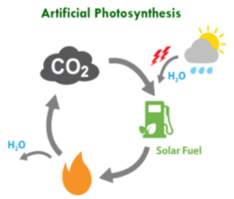
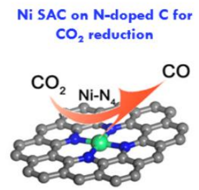
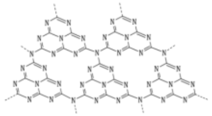
Introduction





Graphitic Carbon Nitride (g-C_3N_4)



- These catalysts were designed to capture and reduce CO_2 into CO which can be used to synthesize useful chemicals such as hydrogen, methanol, and other liquid fuel products[1]
- Graphitic Carbon Nitride, g-C_3N_4, is an ideal semi-conductor since it's cheap, good at absorbing light in the visible range (400-800nm), and it has an ideal bandgap of 2.7eV[2]
- Cobalt Single Atom Catalysts (SACs) complexed on g-C_3N_4 have high selectivity towards CO production

<u>Goal</u>: Determine how heat exfoliations impact the surface area, Cobalt SAC loading, and activity of catalysts synthesized from exfoliated g-C_3N_4

Experimental

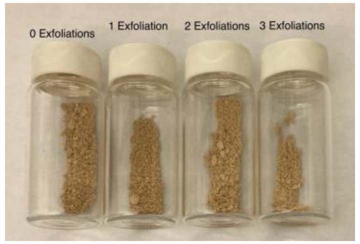
<u>Graphitic C3N4 Synthesis</u>:
Bake 20g Urea with 20mg dextrose at 600°C for 4h[2]

<u>Heat Exfoliation Procedure</u>:
Re-bake the C_3N_4 samples at 600°C for 1h[3]

<u>Cobalt Loading</u>:
Combine 100mg of C_3N_4, 7.5mL Acetonitrile, and 1.25mg of Cobalt, sonicate, stir for 30min, add 65uL of Triethylamine, stir for 30 min, microwave at 80°C for 2h, centrifuge, rinse solids with chloroform, methanol, and acetonitrile, and air dry[4]

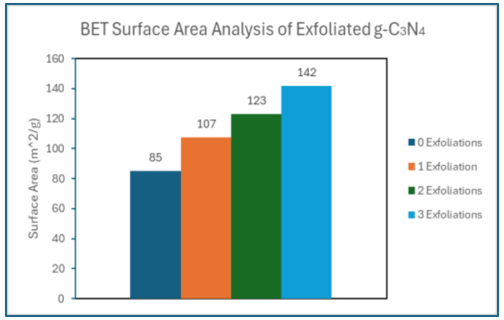
<u>Photocatalysis</u>:
Mix 1mg of catalyst with 4mL of 4:1 Acetonitrile : Triethanolamine, sonicate, bubble with CO_2 for 20min, stir in front of a 200mW/cm^2 lamp with a water filter, and sample the head gas every 30min for 2h for GC analysis[4]

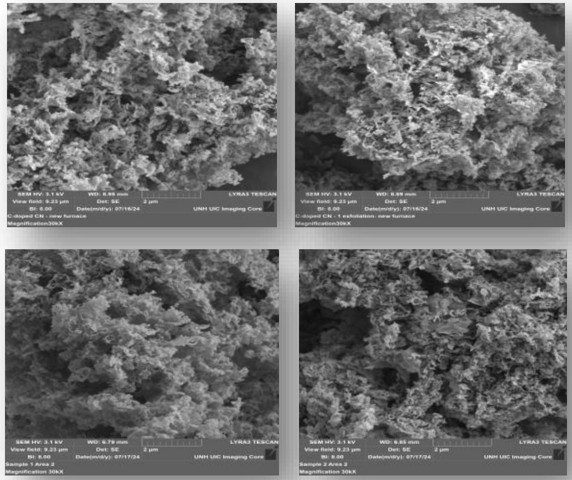
Results



The material's ability to absorb light in the visible spectrum remained unchanged after multiple exfoliations, as can be seen from the UV-Vis data.

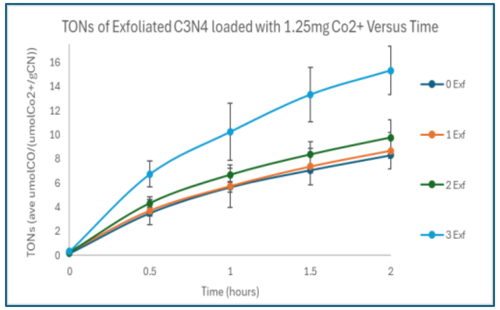
The BET data displays that samples with more exfoliations saw a significant increase in surface area





SEM images of C_3N_4 with 0, 1, 2, & 3 exfoliations (L-R, top-bottom) at 30kx magnification are shown at left

The more exfoliated C_3N_4 samples had more plate-like flakes



The catalysts with more exfoliations appeared to more effectively break down CO_2 into CO

The jump from exfoliations 1 & 2 to 3 & 4 saw a significant increase in Co^{2+} loading

Discussion

The cheap and simple act of re-baking the C_3N_4 before loading it Cobalt SACs has the potential to increase the C_3N_4's surface area, increase its loading potential, and yield more effective catalysts

More successive exfoliations should be performed on a larger batch of C_3N_4 to determine whether the surface area could be further increased by a 4th or 5th exfoliation

In future research, the percent yield should be tracked more thoroughly to determine how much material is lost in each bake

Acknowledgements

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References

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