Unveiling Molecular Affinity: Binding of N-Heterocyclic Carbenes on Carbon Powders

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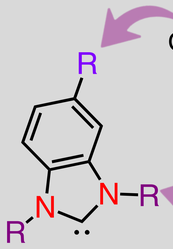
New surface alert!

N-Heterocyclic Carbene (NHC) chemistry has made significant advancements in **applications on gold surfaces.** This project is focused **on carbon** surfaces **due to their stability, reactivity, and unique physical properties,** which can be enhanced by NHC chemistry.

N-Heterocyclic carbenes (NHC)



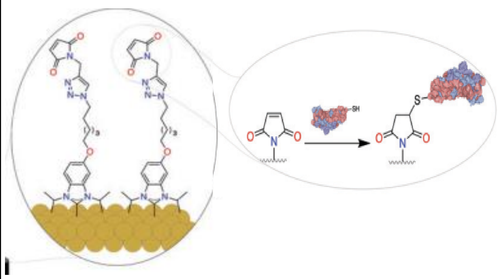
Control surface properties via backbone functionalization

Wingtips contribute to packing and orientation of NHC on surfaces

strong sigma donating ability

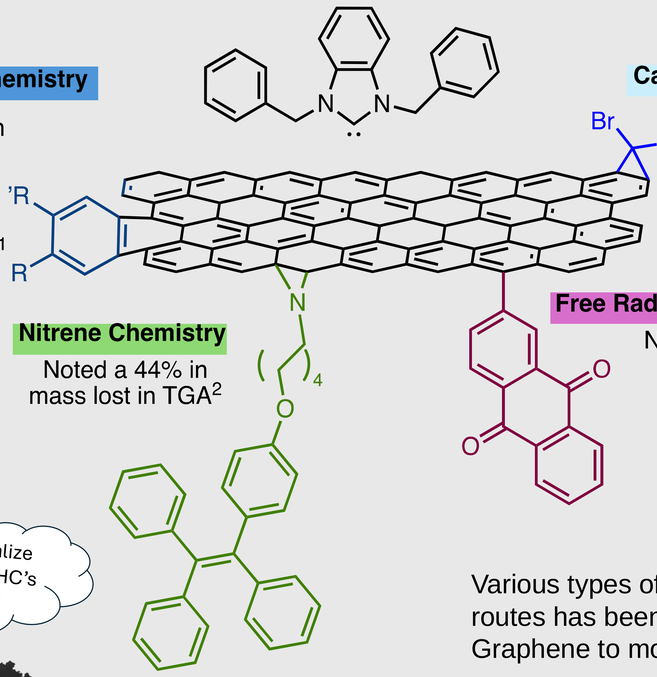
Previous work

Toll-like receptors have been used on the backbone of NHC's that was installed on gold.[5]



Reported functionalization routes on Graphene

N-Heterocyclic Carbene Chemistry



Diels Alder Chemistry

Noted a 36% in mass lost via thermal gravimetric analysis (TGA)[1]

Carbene Chemistry

Noted a 30% in mass lost in TGA.[3]

Nitrene Chemistry

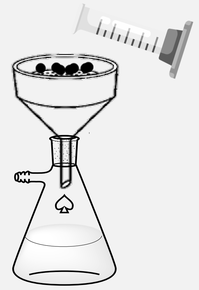
Noted a 44% in mass lost in TGA[2]

Free Radical Chemistry

Noted a 7% in mass lost in TGA[4]

Various types of functionalization routes has been employed on Graphene to modify its properties

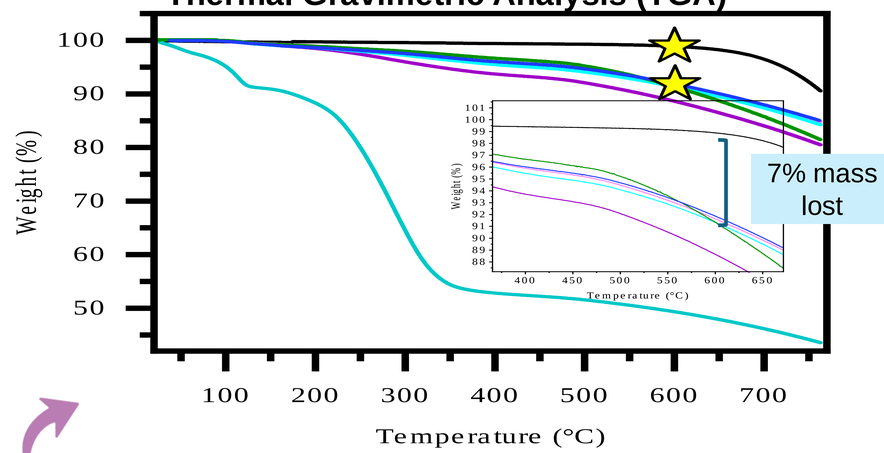
Characterization



Functionalized carbon powders undergo subsequent washing steps to remove unreacted NHC's

- Vulcan XC-72
- First 20 mL wash with dichloromethane
- Second 20 mL wash with dichloromethane
- Third 20 mL wash with dichloromethane
- Fourth 20 mL wash with dichloromethane
- Fifth 20 mL wash with dichloromethane
- Sixth 20 mL wash with dichloromethane

Thermal Gravimetric Analysis (TGA)



Carbon surface properties

- Heterogeneous
- Varying acidic/basic functional groups
- Varying surface area
- Varying porosity

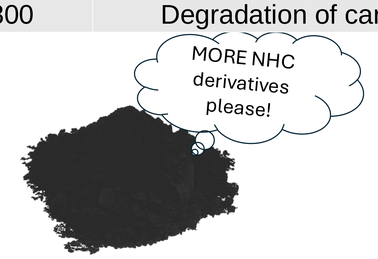
Benefits of using carbon

- highly stable
- conductive
- naturally abundant

Challenge

Carbon is limited by characterization methods to prove functionalization on surfaces. There is also limited understanding of whether functional groups are physiosorbed or chemisorbed.

Temperature (℃)	Mass lost
0-100	Residual organic solvents
200-350	Residual NHC starting material
400-600	Degradation of grafted NHC
600-800	Degradation of carbon



XPS study

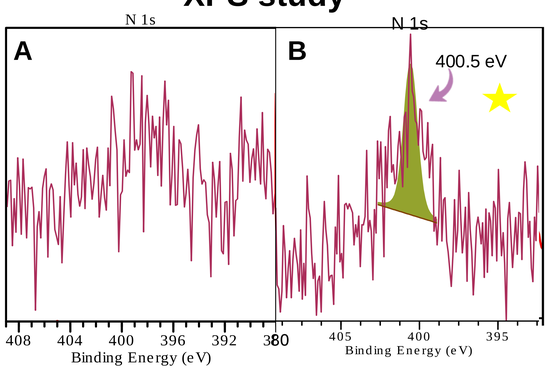
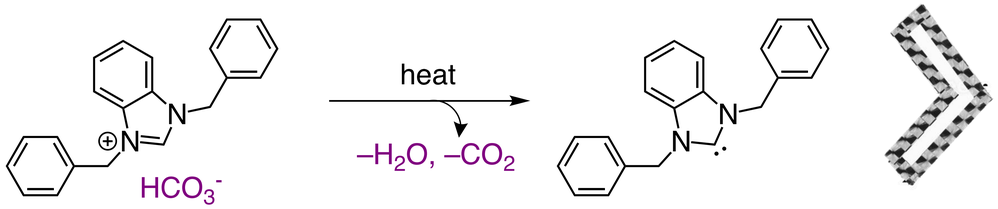
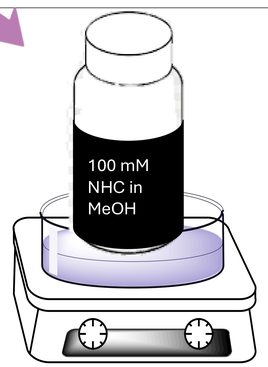


Figure 2. a) carbon powder b) 100 mM NHC deposited on carbon powder

Solution deposition onto carbon powder



Heated at 60℃ and stirred for 48 h



Summary

Dibenzyl benzimidazolium carbonate salts can be covalently added to the surface, and further optimization is being explored.

Future work

- ☆ increase degree of functionalization via exfoliation by sonication and optimizing NHC deposition on oxidized carbon
- Solid-state NMR study
- TOF-SIMS study

References 1) *Chem. Commun.* **2010**, *46*, 7340–7342.2) *RSC Adv.*, **2012**, *2*, 7042–7047. *3) Appli. Surf. Sci.* **2014**, *311*, 377–383. 4) *Carbon.* **2017**, *111*, 83–93. 5) *Chem. Commun.* **2021**, *57*, 8421–8424.