Iron Complexes Anchored onto Magnetically Separable Graphene Oxide Sheets: An Excellent Catalyst for the Synthesis of Dihydroquinazoline-Based Compounds

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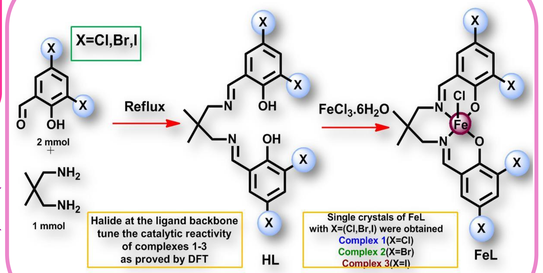
Introduction

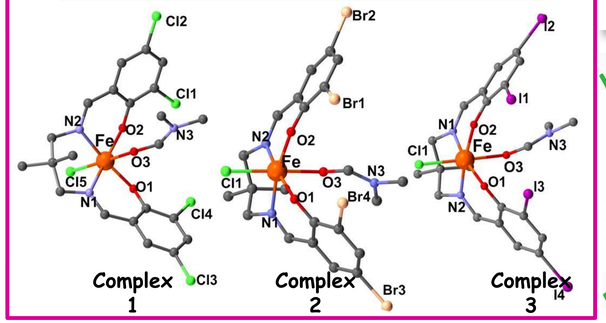
Synthesis of dihydroquinazoline based compounds catalyzed by magnetically separable Graphene oxide nanosheets upholding Iron Schiff base complex (GO@Fe_3O_4@APTES@FeL (GOTESFe).

➢Homogeneous catalysts even though they are highly efficient are difficult to separate from reaction media and hence they cannot be reused for further cycles.

Synthesis of complexes/Homogeneous catalysts



Crystal Structure of the Iron Complexes



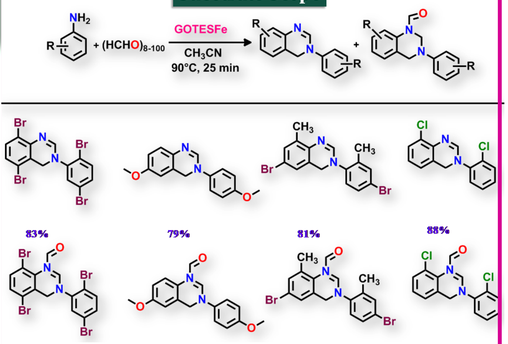
Additionally this present work adheres to SDG 12 (Responsible consumption and production)

✓GOTESFe being a magnetically separable nanoparticle paves way for facile separation from the reaction mixture by a simple application of magnet. Also minimizing catalyst loss, energy consumption, and saves much time in achieving catalyst recovery

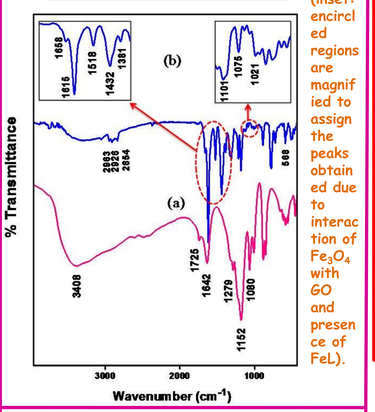
GOTESFe being a magnetically separable nanoparticle paves way for facile separation from the reaction mixture by a simple application of magnet. Usage of such a magnetically separable catalyst also minimises catalyst loss, energy consumption, and saves much time in achieving catalyst recovery

✓We have avoided the use of precious metals such as Au, Ag or Pt and have used alternative environment friendly transition metal Iron

Substrate Scope

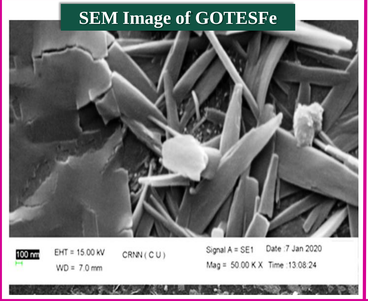


FTIR spectrum of (a) GO (b) GOTESFe



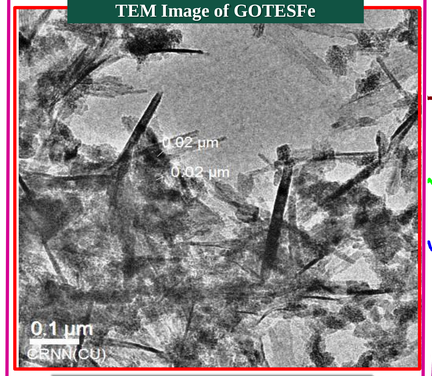
(inset: encircled regions are magnified to assign the peaks obtained due to interaction of Fe_3O_4 with GO and presence of FeL).

SEM Image of GOTESFe



Characterization of GOTESFe

TEM Image of GOTESFe



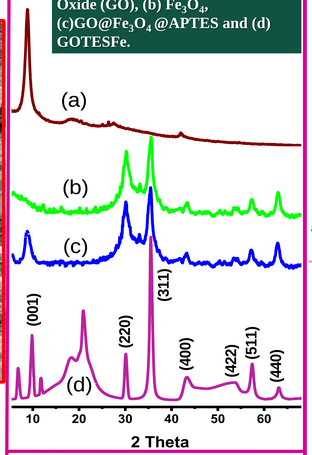
Importance of Dihydroquinazolines

❑These N-heterocycles are the essential infrastructure for relevant drug molecules such as geftinib and tarceva.

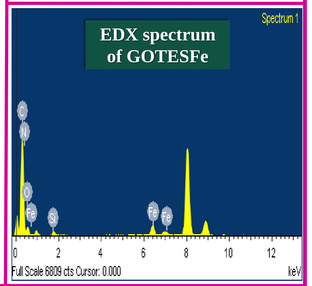
❑Multi substituted quinazoline motifs have profound importance in applications like antibacterial, anticancer, antiviral, antiinflammatory activities, antimalarial activities.

❑ Additionally, dihydroquinazoline derivatives are known to be potential inhibitors of the trypanothione reductase (TryR).

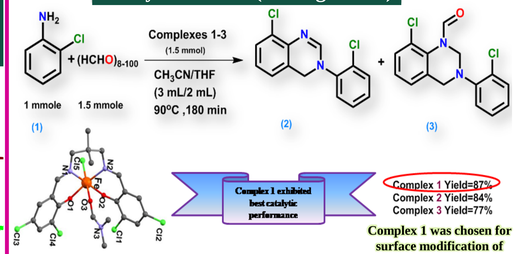
PXRD spectrum of (a) Graphene Oxide (GO), (b) Fe_3O_4, (c)GO@Fe_3O_4 @APTES and (d) GOTESFe.



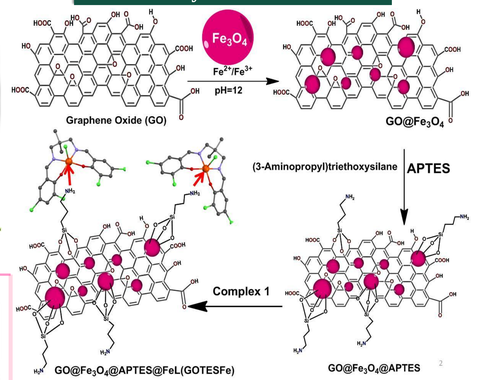
EDX spectrum of GOTESFe



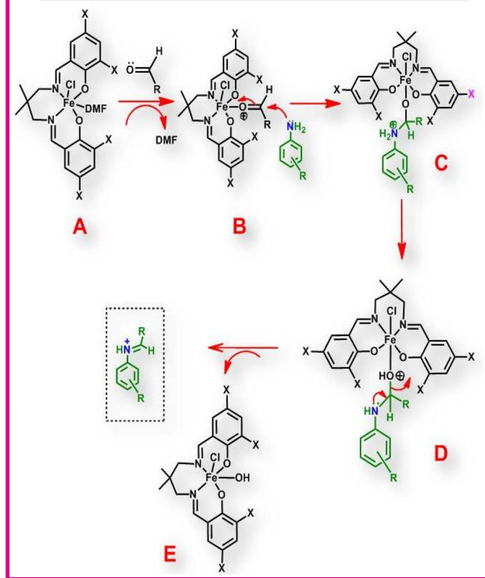
Catalytic reaction(Homogeneous)



Scheme for synthesis of GOTESFe



Most plausible Mechanism for dihydroquinazoline synthesis



Structures	Relative energy	
Complex-1/2/3 +CH_2-O + Aniline	0.0 / 0.0 / 0.0	
A-1/2/3	0.2 / 0.4 / 0.3	
B-1/2/3(Cl / Br / I)	1.8 / 1.9 / 2.0	
TS-BC-1/2/3	**20.7** / **21.0** / **21.3**	
C-1/2/3	-8.2 / -8.1 / -8.2	
D-1/2/3 + E	14.3 / 14.9 / 15.2	

DFT Calculations

DFT calculations helped in rationalising the role of halide substituent in the ligand backbone as a relevant factor in the catalytic superiority of complex 1 over complexes 2 and 3 for the synthesis of the dihydroquinazoline derivatives.

Conclusion

✓Iron(III) Schiff base complexes varying the halide identity in the ligand backbone were used as catalysts for dihydroquinazoline synthesis

✓ Complex 1 excelled among 3 complexes (rationalised by DFT)

✓Complex 1 has been transformed into heterogeneous magnetically separable catalyst by immobilizing it onto GO@Fe_3O_4@APTES resulting in the formation of GOTESFe, which also displayed superior catalytic activity and could be reused up to five cycles

➢GOTESFe is magnetically separable and can be separated after each catalytic cycle using a magnet and can be reused.

References

(1)Maleki et. al. J Porous Mater.,2018, *25*, 1789–1796 (2)Wan et. al.. Tetrahedron 2010,66, 3405–3409.

Acknowledgments

University of Calcutta ,CRNN for providing instrumentation facilities

For Further Reading:- Chakraborty et. al. ACS Appl. Mater. Interfaces,2020, 12, 34, 38530-38545