Dedicated to.....

My beloved parents, brother and my husband

Declaration

I declare that the thesis entitled "**ORGANIC TRANSFORMATIONS USING NOVEL CATALYTIC SYSTEM**" has been prepared by me under the guidance of Dr. Pranab Ghosh, Professor of Chemistry, University of North Bengal. No part of this thesis has formed the basis for the award of any degree or fellowship previously.

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I certify that **Ms. Puja Basak** has prepared the thesis entitled "**ORGANIC TRANSFORMATIONS USING NOVEL CATALYTIC SYSTEM**" for the award of Ph.D. degree of the University of North Bengal, under my supervision. She has carried out the research work at the Department of Chemistry, University of North Bengal. No part of this thesis has formed the basis for the award of any degree or fellowship previously.

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Preface

Scientific interest on graphene and its derivatives in heterogeneous catalysis has grown dramatically over the past several years. The role and the advantages of graphene and chemically derived graphene (CDG) as heterogeneous catalyst cannot be overlooked. Among the heterogeneous catalytic support graphene and its derivatives has been considered as the most promising area of research. Regarding heterogeneous catalyst, various polymeric supports like, zeolite, silica, polymers etc are similarly very important for immobilization of metals as well as other catalytic application. This research work mainly covers the metal-free heterogeneous catalysis, although reactions involving metal-composite catalysts are also described here.

Chapter I is divided into three sections; Section A gives a detailed discussion on graphene oxide (GO) and its application in organic synthesis. In the Section B graphene oxide (GO) used as efficient catalyst for the one-pot synthesis of 3,5-disubstituted 1,2,4-oxadiazoles using robust solid acid catalyst graphene oxide (GO). In Section C from the same chapter shows graphene oxide (GO) as a solid acid catalyst for the synthesis of 2,4,6-triarypyridines from benzaldehydes, acetophenones, and ammonium acetate.

Chapter II is divided into two sections; Section A deals with the aqueous mediated metal-composite catalyzed C–C cross coupling reaction. Section B depicts SuzukiMiyaura and Mizoroki-Heck C–C cross-coupling reaction using low palladium loaded graphene oxide-polymer (GO-PMMA-Pd) composite catalyst under ligand free condition.

Chapter III is divided into two sections. Section A shows the synthesis of substituted isoxazoles using graphene oxide (GO) as metal-free and

environmental friendly catalyst. Section B deals with the one-pot four-component synthesis of substituted pyrazoles in presence of efficient carbocatalyst GO.

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1) Convenient one-pot synthesis of 1,2,4-oxadiazoles and 2,4,6-triarylpyridines using graphene oxide (GO) as a metal-free catalyst: Importance of dual catalytic activity
   Puja Basak, Sourav Dey,and Pranab Ghosh
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5) Graphene Oxide Catalyzed One-pot Synthesis of Pyrimido [4, 5-b] quinolinone-2, 4-diones and their Biological Evaluation
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7) Green Organocatalysis
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8) A design for convenient and greener root towards one pot multi-component synthesis of 7-aryl/heteroaryl substituted pyrano[3,2-c:5,6-c']dichromene-6,8-dione and 7-aryl/heteroaryl substituted chromeno[4,3-d]pyrido[1,2-a]pyrimidinone derivatives using rice husk based heterogeneous catalyst
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Asian Journal of Green Chemistry,
(*accepted*)

9) Catalytic applications of graphene oxide towards the synthesis of bioactive scaffolds through the formation of carbon-carbon and carbon-heteroatom bonds, Green bond forming reactions: carbon-carbon and carbon-heteroatom
Rabindra Nath Singha, **Puja Basak**, and Pranab Ghosh
(*communicated*).

10) Metal-composite catalyzed C-C coupling reactions in wateraqueous mediated heterogeneous catalysis
Puja Basak, and Pranab Ghosh
(*communicated*).

Poster presentation

1) An effective low Pd loading Graphene oxide–polymer supported catalyst for aqueous Suzuki and Heck C-C cross coupling reaction, **Puja Basak**, Pranab Ghosh, National Seminar on Frontiers in Chemistry 2017-2018 organized by Department of Chemistry, University of North Bengal and sponsored by UGC-NEW DELHI, held at University of North Bengal, Darjeeling, September 14, 2017.

2) Explorative studies on Graphene oxide (GO) as an efficient carbocatalyst: synthesis of 3,5 disubstituted 1,2,4 oxadiazole, **Puja Basak**, Pranab Ghosh, International Seminar on Frontiers in Chemistry 2018 organized by Department of Chemistry, University of North Bengal and CRSI North Bengal Local Chapter held at University of North Bengal, Darjeeling, August 27, 2018.

3) A novel graphene oxide (GO) catalysed one-pot synthesis of substituted pyridines, **Puja Basak**, Pranab Ghosh, National Seminar on Frontiers in Chemistry 2018 organized by Department of Chemistry, University of North Bengal and CRSI North Bengal Local Chapter held at University of North Bengal, Darjeeling, August 22, 2019.

Abbreviation

2D	Two dimensional
GO	Graphene oxide
AGO	Amide functionalized graphene oxide
rGO	Reduced graphene oxide
SGO	Sulfonated graphene oxide
tert-BuOK	Potassium tert-butoxide
H_2O_2	Hydrogen peroxide
CDC	Cross-dehydrogenative coupling
MCR	Multicomponent reaction
NPs	Nanoparticles
XPD	X-raydiffraction spectroscopy
XPS	X-ray photoelectron spectroscopy
IR	infrared spectroscopy
TEM	Transmission electron microscopy
TGA	Thermogravimetric analysis
HMF	5-hydroxymethylfurfural
DHPM	Dihydropyrimidone
SRGO	Sulfonated reduced graphene oxide
EDS	Energy dispersive spectroscopy
PTSA	p-Toluenesulfonic acid
DMF	*N,N*-Dimethyl formamide
DMSO	Dimethyl sulfoxide
NBS	N-Bromosuccinimide
DBU	1,8-diazabicyclo[5.4.0]undec-7-ene
HBTU	*N,N,N′,N′*-Tetramethyl-*O*-(1*H*-benzotriazol-1-yl)uronium hexafluorophosphate
DIEA	N,N-Diisopropylethylamine
PS	Polystyrene
BEMP	2-*tert*-Butylimino-2-diethylamino-1,3-dimethylperhydro-1,3,2-diazaphosphorine
MW	Microwave

TBTU	*N,N,N′,N′*-Tetramethyl-*O*-(benzotriazol-1-yl)uronium tetrafluoroborate
DCC	*N,N′*-Dicyclohexylcarbodiimide
EDC	*N*-(3-Dimethylaminopropyl)-*N′*-ethylcarbodiimide hydrochloride
HOBt	1-Hydroxybenzotriazole
TLC	Thin layer chromatography
TEA	Triethylamine
SEM	Scanning electron microscope
EDX	Energy-dispersive X-ray spectroscopy
ppm	Parts per milion
HRMS	High resolution mass spectroscopy
TMS	tetramethylsilane
PFPAT	Pentafluorophenylammonium triflate
IL	Ionic liquid
rt	Room temperature
n-TSA	Nano titania-supported sulfonic acid
MNP	Magnetic nanoparticles
FT-NMR	Fourier Transform- Nuclear magnetic resonance
CDG	Chemically derived graphene
Nf	Nafion
PCA	1-Pyrenecarboxylic acid
DMA	*N,N*-Dimethylacetamide
NMP	*N*-Methyl-2-Pyrrolidone
PMMA	Poly (methyl methacrylate)
BZP	Benzoyl peroxide
ICP-AES	Inductively Coupled Plasma-Atomic Emission Spectrometers
PPI	Potassium phthalimide
TCNE	Tetracyanoethylene
HPA	Heteropolyacid
DCM	Dichloromethane
THF	Tetrahydrofuran
br	broad
d	Doublet

Hz	Hertz
m	Multiplet
s	Singlet
t	Triplet
min	Minute
mg	Milligram
^{o}C	Degree Celsius
h	Hour
mL	milliliter
wt %	Weight percent
PPh3	Triphenylphosphine
TBAB	Tetrabutylammonium bromide
SDS	Sodium dodecyl sulfate
TPAOH	Tetrapropylammonium hydroxide
Pd@zeolite	Zeolite-supported Pd catalyst
CS	Chitosan
MPTA	Poly-triallylamine